Tensor-Train Split-Operator KSL (TT-SOKSL)

Method for Quantum Dynamics Simulations

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Abstract

Numerically exact simulations of quantum reaction dynamics, including non-adiabatic effects in excited electronic states, are essential to gain fundamental insights into ultrafast chemical reactivity and rigorous interpretations of molecular spectroscopy. Here, we introduce the tensor-train split-operator KSL (TT-SOKSL) method for quantum simulations in tensor-train (TT)/matrix product state (MPS) representations. TT-SOKSL propagates the quantum state as a tensor train using the Trotter expansion of the time-evolution operator, as in the tensor-train split-operator Fourier transform (TT-SOFT) method. However, the exponential operators of the Trotter expansion are applied using a rank adaptive TT-KSL scheme instead of using the scaling and squaring approach as in TT-SOFT. We demonstrate the accuracy and efficiency of TT-SOKSL as applied to simulations of the photoisomerization of the retinal chromophore in rhodopsin, including non-adiabatic dynamics at a conical intersection of potential energy surfaces. The quantum evolution is described in full dimensionality by a time-dependent wavepacket evolving according to a two-state 25-dimensional model Hamiltonian. We find that TT-SOKSL converges faster than TT-SOFT with respect to the maximally allowed memory requirement of the tensor-train representation and
better preserves the norm of the time-evolving state. When compared to the corre-
sponding simulations based on the TT-KSL method, TT-SOKSL has the advantage of
avoiding the need of constructing the matrix product state Laplacian by exploiting the
linear scaling of multidimensional tensor train Fourier transforms.

1 Introduction

Simulations of quantum phenomena in chemical and biological systems typically require
time-dependent methods. For example, photoinduced reactions as well as processes
that involve energy transfer, electron transfer, simulations of molecular spectroscopy, and
coherent control require rigorous descriptions of quantum effects, including tunneling,
interference, entanglement, and non-adiabatic dynamics. Simulations in the time-
dependent picture require integration of the time-dependent Schrödinger equation (TDSE)
explicitly, which can be efficiently performed for small molecular systems for example by
using the split-operator Fourier transform (SOFT) method which is a numerically exact
method. However, SOFT is limited to systems with very few degrees of freedom (DOF)
(i.e., molecular systems with less than 4 or 6 atoms), since it is based on a full basis
set representation requiring storage space and computational effort that scale exponentially
with the number of coupled DOFs. Other numerically exact quantum dynamics methods in-
clude the Chebyshev polynomial expansion method and methods based on the Krylov
expansion.

The exponential scaling problem has motivated the development of a variety of methods
based on truncated basis sets. Some of these methods employ Gaussian coherent states,
such as the method of coupled coherent states, the multiple-spawning method, and the matching-pursuit algorithm. The Multi-Configurational Time-Dependent Hartree
(MCTDH) method groups DOFs into “particles” represented in a DVR basis and has been implemented for efficient calculations in terms of the so-called multilayer MCTDH
method. However, determining how exactly the basis should be truncated or how to group
DOFs into particles can be difficult and relies on approximations.\textsuperscript{32} It is also noted that the MCTDH equations of motion involve ill-conditioned matrices which lead to numerical instabilities during propagation and fundamentally restrict the propagation time step for any standard integrator.\textsuperscript{33}

In earlier work, we have introduced the so-called tensor-train (TT) split-operator Fourier transform (SOFT) method (TT-SOFT)\textsuperscript{35} that allows for rigorous simulations of multidimensional nonadiabatic quantum dynamics. TT-SOFT represents the time-dependent wave function as a dynamically adaptive TT and evolves it by recursively applying the time-evolution operator as defined by the Trotter expansion. Exploiting the efficient TT implementation of multidimensional Fourier transforms, the TT-SOFT algorithm applies the Trotter expansion of the time-evolution operator using exponential operators in tensor-train format generated by the scaling and squaring method. The accuracy and efficiency of the TT-SOFT method were demonstrated as applied to the propagation of 24-dimensional wave packets describing the $S_1/S_2$ non-adiabatic dynamics of interconversion of pyrazine after UV photoabsorption. Here, we introduce the tensor-train split-operator KSL (TT-SOKSL) method, which, although it is also based on the Trotter expansion of the time-evolution operator, has the advantage of avoiding the computational bottleneck of scaling and squaring by applying the exponential operators according to a rank-adaptive version of the TT-KSL solver.\textsuperscript{36–38}

The TT-KSL algorithm\textsuperscript{36,37} is a TT implementation of the dynamical low-rank approximation method for evolution of time-dependent matrices.\textsuperscript{38} Rather than generating a low-rank approximation by rounding after generating a high-rank solution, TT-KSL implements an orthogonal projection onto a low-rank manifold. It has been applied in conjunction with the Dirac-Frenkel Time-Dependent Variational Principle (TDVP), as demonstrated in applications to simulations of pyrazine,\textsuperscript{43} the Fenna-Matthews-Olson (FMO) complex,\textsuperscript{44} and singlet fission in molecular dimer and Perylene-Bisimide aggregates.\textsuperscript{45} The method has typically been carried out as an effective matrix-vector multiplication scheme in the occupation number representation using the kinetic operator matrix in TT format. When implemented
in the position grid representation, the kinetic operator requires a finite-difference approximation\cite{10} or implementations based on the Fourier Grid Hamiltonian (FGH)\cite{11,12} or similar Discrete Variable Representation (DVR) methods.\cite{50,51} The TT-SOKSL method avoids matrix-vector multiplication schemes by exploiting the diagonal representation of the kinetic operator in momentum space to evolve the quantum state by vector-vector multiplication. TT-SOKSL thus combines the simplicity of TT-SOFT and the advantages of a projector-splitting integrator to implement the exponential operators of the Trotter expansion in diagonal form.

We demonstrate the capabilities of TT-SOKSL as applied to simulations of non-adiabatic quantum dynamics. We focus on the photoisomerization process of the retinal chromophore in rhodopsin as described by a two-state 25-mode model Hamiltonian.\cite{52} The model system is ideally suited for comparisons to previous calculations based on TT-SOFT and the MCTDH methods.

2 Methods

2.1 Potential energy surface

We simulate the nuclear and electronic dynamics of the photoisomerization of the retinal molecule (Figure 1) to explore the capabilities of the TT-SOKSL method as compared to TT-SOFT and MCTDH. The model Hamiltonian\cite{52,53} consists of the vibronically coupled $S_0$ and $S_1$ diabatic potential energy surfaces (PES). These are 25-dimensional PESs parametrized by

Figure 1: 11-cis/all-trans photoinduced isomerization of the retinyl chromophore in visual rhodopsin.
the resonance Raman active modes of the retinyl chromophore in rhodopsin. Two modes are identified as the large-amplitude primary reactive modes, which correspond to the C_{11}=C_{12} torsion and the carbon-carbon stretching mode of the conjugated double bonds (Figure 2). The other 23 modes are modeled as harmonic oscillators linearly coupled to the excited electronic state with frequencies and equilibrium positions parametrized by the experimental

Figure 2: Diabatic potential energy surfaces of the rhodopsin ground (S_0) and first excited (S_1) electronic states as a function of the two large amplitude coordinates, the torsion dihedral angle coordinate \( \theta \) of the C_{11}C_{12} bond and the stretching coordinate \( q_c \) of the conjugated polyene chain. Black lines connect cis/trans wells with the cis/trans conformations of the molecule. The orange arrow indicates the initial photo-excitation (\( \lambda = 500 \) nm) to the \( S_1 \) surface.
resonance Raman spectrum, as follows:

\[ H = H_S + H_B, \]

\[ H_S = \sum_{n,m=0}^{1} |\psi_{el}^n(T_s\delta_{mn} + V_{mn})\rangle \langle \psi_{el}^n|, \]

\[ T_s = -\frac{\Omega_c}{2} \frac{\partial^2}{\partial q_c^2} - \frac{1}{2I} \frac{\partial^2}{\partial \theta^2}, \]

\[ V_{00} = \frac{W_0}{2}(1 - \cos\theta) + \frac{\Omega_c}{2} q_c^2, \]

\[ V_{11} = E_1 - \frac{W_1}{2}(1 - \cos\theta) + \frac{\Omega_c}{2} q_c^2 + \kappa q_c, \]

\[ V_{10} = V_{01} = \lambda q_c, \]

\[ H_B = \sum_{n=0}^{1} \sum_{i=1}^{23} |\psi_{el}^n(\omega_i)\rangle \left( \frac{\omega_i}{2} \left( \frac{\partial^2}{\partial q_i^2} + q_i^2 \right) + \kappa_i q_i \delta_{ni} \right) \langle \psi_{el}^n|. \]

Here, \( \Omega_c = 1532 \text{ cm}^{-1} \), with remaining parameters (in eV): \( 1/I = 4.84 \times 10^{-4}, E_1 = 2.48, W_0 = 3.6, W_1 = 1.09, \kappa = 0.1, \lambda = 0.19 \). Parameters of the other vibrational modes \( \{\kappa_i, \omega_i\} \) as well as our Python codes for reproducing all of the results reported in this paper are available at https://github.com/NingyiLyu/TTSOKSL. We note that a new set of parameters has been recently reported\(^{54,55} \) to better match experimental findings.\(^{54,55} \) Nevertheless, our calculations are based on the original set of parameters to allow for comparisons to earlier studies.\(^{50,50} \)

### 2.2 Initial Conditions

We initialize the wavepacket as a Gaussian on the \( S_1 \) excited state, centered at the equilibrium position of the vibrational modes in the ground electronic state, to simulate the light-induced \( S_1 \leftarrow S_0 \) vertical transition:

\[ \psi(x_1, \ldots, x_{25}; 0) = \prod_{j=1}^{25} (\sigma_j^2 \pi/2)^{-1/4} e^{-x_j^2/\sigma_j^2}, \]
where $\sigma_1 = 0.15228$ a.u., $\sigma_j = \sqrt{2}$ a.u., with $j = 2$–25. The wavepacket then evolves according to the coupled $S_1/S_0$ potential energy surfaces, which partitions the population between the cis/trans conformations in the ground and excited states as it reaches configurations close to the conical intersection. The isomerization quantum yield (i.e., trans:cis population ratio) is computed by integrating the trans population as defined by configurations with $|\theta| > \pi/2$ where, according to Eq. (1), $\theta$ is the dihedral angle about the C11–C12 bond.

### 2.3 Tensor-train decomposition

TT-SOKSL relies on the tensor-train (TT) format, also called matrix product states (MPS) with open boundary conditions, recently explored for the development of methods for quantum dynamics and global optimization.

The TT format of an arbitrary $d$-dimensional tensor $X \in \mathbb{C}^{n_1 \times \cdots \times n_d}$ involves a train-like matrix product of $d$ 3-mode tensors $X_i \in \mathbb{C}^{r_i-1 \times n_i \times r_i}$ with $r_0 = r_d = 1$, so any element $X(j_1, \ldots, j_d)$ of $X$ can be evaluated, as follows:

$$X(j_1, \ldots, j_d) = \sum_{a_0=1}^{r_0} \sum_{a_1=1}^{r_1} \ldots \sum_{a_d=1}^{r_d} X_1(a_0, j_1, a_1)X_2(a_1, j_2, a_2)\ldots X_d(a_{d-1}, j_d, a_d), \quad (3)$$

or equivalently in matrix product notation,

$$X(j_1, \ldots, j_d) = X_1(j_1)\ldots X_d(j_d), \quad (4)$$

where $X_i(j) \in \mathbb{C}^{r_i-1 \times r_i}$ is the $j$-th slice of tensor core $X_i$. The TT-rank is defined in terms of the vector $r = \{r_0, r_1, \ldots, r_{d-1}, r_d\}$ introduced by Eq. (3). We note that the resulting TT representation of $X$, with $n_1 = n_2 = \cdots = n_d = n$ and $r_1 = r_2 = \cdots = r_{d-1} = \tilde{r}$, requires only $n \times d \times \tilde{r}^2$ data points, which bypasses the usual exponential $n^d$ number of elements required by the full-rank representation.

TT-matrices $M \in \mathbb{C}^{(n_1 \times n_1) \times (n_2 \times n_2) \times \cdots \times (n_d \times n_d)}$, also called matrix-product-operators (MPO),
are defined with analogous format,

$$M(l_1, \ldots, l_d; j_1, \ldots, j_d) = M_1(l_1, j_1) \ldots M_d(l_d, j_d), \quad \text{with} \quad M_i(l_i, j_i) \in \mathbb{C}^{rM_{i-1} \times rM_i}. \quad (5)$$

Therefore, TT-matrices $M$ operate core-by-core on TT-vectors $X \in \mathbb{C}^{n_1 \times \ldots \times n_d}$, as follows:

$$MX(l_1, \ldots, l_d) = \sum_{j_1, \ldots, j_d} M(l_1, \ldots, l_d; j_1, \ldots, j_d)X(j_1, \ldots, j_d),$$

$$= \sum_{j_1, \ldots, j_d} (M_1(l_1, j_1) \otimes X(j_1)) \ldots (M_d(l_d, j_d) \otimes X(j_d)). \quad (6)$$

### 2.4 TT-SOKSL

#### 2.4.1 Split-operator propagator

The TT-SOKSL method integrates the time-dependent Schrödinger equation:

$$\dot{\Psi}(t) = -\frac{i}{\hbar} \hat{H} \Psi(t) \quad (7)$$

where $\hat{H} = \hat{T} + \hat{V}$ is the Hamiltonian of the system and $\Psi(t)$ is the time-dependent state. Equation (7) can be integrated to second-order accuracy by using the Trotter (Strang-splitting) approximation, as in the SOFT method.\textsuperscript{17-19} For each integration time-step $\tau = t_{k+1} - t_k$, we evolve the state from $\Psi(t_k)$ to $\Psi(t_{k+1}) = \Psi(t_k + \tau)$, as follows. First, we obtain $\Psi_1(t_k + \tau)$ by integrating the equation,

$$\dot{\Psi}_1(t) = -\frac{i}{2\hbar} \hat{V} \Psi_1(t), \quad (8)$$

with $\Psi_1(t_k) = \Psi(t_k)$. Then, we obtain $\Psi_2(t_k + \tau)$ by integrating the equation,

$$\dot{\Psi}_2(t) = -\frac{i}{\hbar} \hat{T} \Psi_2(t), \quad (9)$$
with $\Psi_2(t_k) = \Psi_1(t_k + \tau)$ and we obtain $\Psi_3(t_k + \tau)$ by integrating the equation,

$$\dot{\Psi}_3(t) = -\frac{i}{2\hbar} \hat{V} \Psi_3(t),$$

(10)

with $\Psi_3(t_k) = \Psi_2(t_k + \tau)$ to obtain $\Psi(t_{k+1}) = \Psi_3(t_k + \tau) + O(\tau^3)$.

TT-SOKSL represents $\Psi(t)$ as a TT-vector and sequentially integrates Eqs. (8)–(10) by using the rank-adaptive implementation of the so-called dynamical low-rank approximation (TT-KSL method, Appendix F). Our rank-adaptive scheme ensures that the TT rank does not limit the accuracy of the propagation. As in the TT-SOFT algorithm, TT-SOKSL exploits the efficient implementation of multidimensional Fourier transforms in TT format. So, Eq. (8) is integrated in the coordinate representation by vector-vector multiplication, using the TT-vector operator $-i\hat{V}/2\hbar$. $\Psi_1(t_k + \tau)$ obtained in TT format is then Fourier transformed (FT), and Eq. (9) is integrated in momentum space also by vector-vector multiplication using the TT-vector $-i\hat{T}/\hbar$. After an inverse Fourier Transform (IFT) of $\Psi_2(t_k + \tau)$ back to the coordinate representation, Eq. (10) is solved akin to Eq. (8) to obtain the time-evolved state, as follows:

$$\Psi(x, t_{k+1}) =$$

$$KSL \left( \tau, -i\hat{V}(x)/2\hbar, \text{FT} \left[ KSL \left( \tau, -i\hat{T}(p)/\hbar, \text{IFT} \left[ KSL \left( \tau, -i\hat{V}(x')/2\hbar, \Psi(x', t_k) \right) \right] \right] \right) \right),$$

(11)

where KSL denotes the rank-adaptive TT-KSL integration substeps.

Eq. (11) shows that the potential and kinetic energy operators are applied as TT-vectors in the coordinate and momentum representations, respectively, which bypasses the need to construct matrix product operators in TT-format as required by the original KSL method.

Each of the Eqs. (8)–(10) can be represented, as follows:

$$\dot{\Psi}(t) = M\Psi(t),$$

(12)
where $\mathbf{M}$ is the diagonal TT-matrix representation of a TT-vector, defined as $\mathbf{M} = -i\mathbf{V}/2\hbar$ for Eqs. (8) and (10) and $\mathbf{M} = -i\mathbf{T}/\hbar$ for Eq. (9), with $\mathbf{V}$ and $\mathbf{T}$ the potential and kinetic energy matrices in coordinate- and momentum-space representations, respectively.

2.4.2 Dynamical Low-Rank Approximation: KSL algorithm

The dynamical low-rank approximation method\cite{72} is an efficient algorithm to obtain an approximate solution of Eq. (12) in the form of a matrix $\mathbf{Y}(t)$ of specified rank $r$. Rather than obtaining a solution with high rank and then truncating it by singular value decomposition (SVD), the dynamical low-rank approximation method integrates the following equation,

$$\dot{\mathbf{Y}}(t) = \mathbf{P}_\mathbf{Y}(\mathbf{M}\mathbf{Y}(t)),$$

(13)

The operator $\mathbf{P}_\mathbf{Y}$ projects $\mathbf{M}\mathbf{Y}(t)$ onto the tangent plane $T_{\mathbf{Y}}\mathcal{M}_r$ (Figure 3) – i.e., the plane tangent to the manifold $\mathcal{M}_r$ of states of rank $r$ at $\mathbf{Y}(t)$. After each propagation time-step $\tau$, the resulting approximate solution $\mathbf{Y}(t + \tau)$ is the state on the manifold $\mathcal{M}_r$ that is closest to the exact higher-rank solution. Appendices A–D provide the derivation of $\mathbf{P}_\mathbf{Y}$ and its implementation according to Eq. (13). An important advantage of the KSL propagation scheme is that it does not require matrix inversion or any kind of regularization scheme as typically implemented in other propagation methods, such as MCTDH.\cite{73}
The TT-KSL method is the tensor-train implementation of the dynamical low-rank approximation. It is based on the following expression of $P_Y$ (with derivation and implementation explained in Appendices E-F):

$$
P_Y(MY) = \sum_{i=1}^{d-1} \text{Ten}_i[(I_{n_i} \otimes P_{\leq i-1})[MY]^{(i)}P_{\geq i+1}-P_{\leq i}[MY]^{(i)}P_{\geq i+1}]+\text{Ten}_d[(I_{n_d} \otimes P_{\leq d-1})[MY]^{(d)}],
$$

where matrices are in bold, with $[MY]^{(i)} \in \mathbb{R}^{n_1 \times \ldots \times n_i \times n_{i+1} \ldots n_d}$ the $i$-th unfolding of the tensor train $MY \in \mathbb{R}^{n_1 \times \ldots \times n_d}$. Here, Ten$_i$ denotes the construction of the tensor train from its $i$-th unfolded matrix, $P_{\leq i-1}$ and $P_{\geq i+1}$ denote partial tensor products of tensor cores with indices $\leq i-1$ and $\geq i+1$, respectively (Appendix D.3). Appendix D provides a detailed description of the tensor-train notation, including left- and right-orthogonalization relative to tensor core $i$, which generates $U_{\leq i}$ and $V_{\geq i}$, with $P_{\leq i} = U_{\leq i}U_{\leq i}^T$ and $P_{\geq i} = V_{\geq i}V_{\geq i}^T$, the orthogonal projectors onto $U_{\leq i}$, and $V_{\geq i}$, respectively.

2.4.3 Projector-splitting integrator

The TT-KSL method integrates the equation $\dot{Y}(t) = P_Y(MY(t))$ by splitting $P_Y$. The specific splitting scheme can be introduced with the following notation. Denoting $P_i^+(MY) =$
\[
\begin{align*}
\text{Ten}_i[(I_n \otimes P_{\leq i-1})[MY]^{(i)}P_{\geq i+1}] , \ P_i^-(MY) &= \text{Ten}_i[-P_{\leq i}[MY]^{(i)}P_{\geq i+1}] \text{ for } i = 1, \ldots, d - 1, \\
\text{and } P_d^+(MY) &= \text{Ten}_d[(I_n \otimes P_{\leq d-1})[MY]^{(d)}],
\end{align*}
\]

we rewrite Eq. (14), as follows:
\[
\dot{Y} = P_Y(MY) = P_1^+(MY) - P_1^-(MY) + P_2^+(MY) - P_2^-(MY) + \cdots - P_{d-1}^-(MY) + P_d^+(MY),
\]

where the right-hand side of Eq. (15) is a sum of \(2d - 1\) terms, which can be implemented as the following sequence of initial value problems on the time interval \([t_0, t_1]\):
\[
\begin{align*}
\dot{Y}_1^+ &= P_1^+(MY_1^+), \quad Y_1^+(t_0) = Y_0 \\
\dot{Y}_1^- &= P_1^-(MY_1^-), \quad Y_1^-(t_0) = Y_1^+(t_1) \\
\vdots \\
\dot{Y}_i^+ &= P_i^+(MY_i^+), \quad Y_i^+(t_0) = Y_{i-1}^-(t_1) \\
\dot{Y}_i^- &= P_i^-(MY_i^-), \quad Y_i^-(t_0) = Y_i^+(t_1) \\
\vdots \\
\dot{Y}_d^+ &= P_d^+(MY_d^+), \quad Y_d^+(t_0) = Y_{i-1}^-(t_1),
\end{align*}
\]

where \(t_1 = t_0 + \tau\) and \(Y_0 = Y(t_0)\) is the initial value that corresponds to the wavefunction before the update \(Y(t_1) \approx Y_d^+(t_1)\). The resulting splitting greatly facilitates the integration of \(\dot{Y}(t) = P_Y(MY(t))\) by sequentially updating core-by-core according to \(\dot{Y}_i^+\) and \(\dot{Y}_i^-\), as shown in Appendix F.

The solution of the equation \(\dot{Y}_i^+ = P_i^+(\hat{A})\) can be written, as follows:
\[
Y_i^+(t_1) = U_{\leq i-1}(t_0)K_i^<(t_1)V_{\geq i+1}^T(t_0),
\]
with
\[
K_i^<(t_1) = e^{(t_1-t_0)W_i}K_i^<(t_0),
\]
where $W_i$ is defined, as follows:

$$W_i K_i^<(t) = (I_{n_i} \otimes U_{\leq i-1}(t_0))(I_{n_i} \otimes \bar{U}_{\leq i-1}(t_0)) \bar{K}_i^<(t) \bar{V}^T_{\geq i+1}(t_0) V_{\geq i+1}(t_0).$$  \hfill (19)

Here, $\bar{U}_{\leq i-1}$ represents the left-unfolding matrices of the first $i-1$ cores of $MY^+$ and $\bar{V}_{\geq i+1}$ represents the right-unfolding matrices of the last $d-i$ cores. $K_i^<(t)$ corresponds to the $i$-th core, obtained by operating the $i$-th core of $M$ (i.e., $M_i$) on $K_i^<(t)$, with proper reshaping (Appendix F.3). The operator exponential $e^{(t_1-t_0)W_i K_i^<(t_0)}$ is efficiently evaluated in the Krylov subspace, as implemented in the EXPOKIT package.\(^{23}\)

The update of core $i$ is completed by integration of the differential equation of motion

$$\dot{Y}_i^{-}(t) = P_i^{-}(MY_i^{-}), \text{ with } Y_i^{-}(t_0) = Y_i^{+}(t_1).$$

The initial state is orthogonalized at core $i$, as follows:

$$Y_i^{-}(t_0)_{(i)} = U_{\leq i-1}(t_0) K_i^<(t_1) V^T_{\geq i+1}(t_0),$$

$$= U_{\leq i-1}(t_0) U_i^<(t_1) S_i(t_0) V^T_{\geq i+1}(t_0),$$

$$= U_{\leq i}(t_1) S_i(t_0) V^T_{\geq i+1}(t_0),$$

where $U_i^<(t_1) S_i(t_0) = K_i^<(t_1)$, with $U_i^<(t_1)$ and $S_i(t_0)$ obtained by QR decomposition of $K_i^<(t_1)$. An approach analogous to Eqs. (17)-(19) can now be applied to update $S_i$ (see Appendix F), giving $Y_i^{-}(t_1)_{(i)} = U_{\leq i}(t_1) S_i(t_1) V^T_{\geq i+1}(t_0)$.

Having updated the first core ($i = 1$), the same procedure is then sequentially applied to update all other cores $i = 2-d$ according to what is called a ‘forward sweep’ update of the tensor, from $Y(t_0)$ to $Y(t_1)$, which is a first-order integrator. Sweeping in reverse (i.e., swapping $1,...,d$ with $d,...,1$ in Eq. (16)) results in an alternative first-order integrator, called a ‘backward sweep’. Combining a forward and a backward sweep with half a time step results in a second-order integrator.\(^{36}\) That second-order KSL integrator is indicated in Eq. (11), as follows:

$$Y(t_0 + \tau) = \text{KSL}(\tau, M, Y(t_0)),$$

\hfill (21)
where $Y(t_0)$ is the state to be updated, $\tau$ is the time-step, and $M$ is defined by either the kinetic or potential energy term of the Hamiltonian according to the corresponding steps of the SOFT propagation defined in Eqs. (8)–(10).

### 2.4.4 TT-SOKSL rank-adaptive scheme

We adapt the rank during each TT-KSL substep of the TT-SOKSL scheme, introduced by Eq. (11), to evolve the state with the minimum rank that does not compromise the accuracy. After obtaining $Y(t_1)$ from $Y(t_0) \in M_r$, the propagation is repeated from an initial state with augmented rank $\tilde{Y}(t_0) \in M_{r+1}$ to obtain $\tilde{Y}(t_1)$, where $\tilde{Y}(t_0)$ is obtained by adding to $Y(t_0)$ a random tensor train of fixed rank (e.g., rank-1) and very small norm (e.g., $10^{-12}$). If the overlap of $\tilde{Y}(t_1)$ and $Y(t_1)$ is sufficiently close to unity, $Y(t_1)$ is used as the time-evolved state. Otherwise, $\tilde{Y}(t_1)$ is used as the initial condition for the next propagation time step, which thus provides rank adaptivity.

### 3 Results

Figure 4 (a) and (b) compare the time-dependent population of the ground state and the trans population ($S_0+S_1$) for a 2-dimensional (2D) model of the cis/trans isomerization of the retinyl chromophore that allows for comparisons of TT calculations to numerically exact full-grid quantum dynamics simulations. The excellent agreement shows that TT-SOKSL produces results in quantitative agreement with numerically exact calculations.

Figure 4 shows that transitions to the ground state and formation of the trans isomer begin at $\sim 80$ fs after photoexcitation. The trans population curve exhibits a rapid growth reaching its maximum at $\sim 180$ fs, where about 70% of the total population is in the trans form, which successfully describes the primary isomerization event for this ultrafast reaction.$^{33}$ That primary event is followed by relaxation to the ground state, as shown by both curves of time-dependent populations that continue to exhibit strong oscillations during the
first ps of dynamics, in agreement with previous studies.\textsuperscript{57,60}

Figure 4(c) shows the comparative analysis of the time-dependent TT-rank for TT-SOKSL, TT-KSL, and TT-SOFT. Clearly, TT-SOKSL allows for efficient propagation when compared to TT-KSL and TT-SOFT. The TT ranks of all three methods grow steadily during the primary isomerization event. TT-SOKSL and TT-KSL are very comparable and reach a rank of about 10, whereas TT-SOFT requires a higher rank for comparable precision. These results show that the KSL algorithm is able to evolve the time-dependent state with a lower-rank representation than an algorithm based on the implementation of the time-evolution operator by scaling and squaring followed by rounding, as implemented by TT-SOFT.
Figure 4: (a) Diabatic trans population of the 2D retinal model. (b) Diabatic ground state population of the 2D retinal model. (c) TT-rank structure of the 2D retinal simulation. eps is the accuracy parameter of TT rounding, described in Ref. and threshold $= \langle Y(t) | Y(t) \rangle - 1$ as described in Sec. 2.4.4.

Figure 5 shows the results of simulations for the 25-dimensional (25D) model, showing the capabilities of TT-SOKSL as applied to simulations of non-adiabatic dynamics in high-dimensional model systems. Figure 5(a) shows the time-dependent trans population ($S_0 + S_1$)
and (b) shows the overall $S_0$ population as they evolve during the first ps of dynamics after photo-excitation to the $S_1$ state. Analysis of the time-dependent populations shows that TT-SOKSL and TT-KSL match closely, whereas TT-SOFT shows some deviations at the longer times. The three methods predict that the main isomerization begins at about $\sim 80$ fs after excitation, reaching a maximum of trans population at $\sim 180$ fs of dynamics. At 300-1000 fs, after isomerization, all three methods predict a smooth decay of the trans population and a smooth increase of the $S_0$ population, in contrast to the strong oscillations observed in the 2D model. In summary, the analysis of population dynamics shown in Figure 5 clearly shows that the results obtained with TT-SOKSL agree very well with those obtained with the state-of-the-art TT-KSL method.

![Figure 5](image)

**Figure 5:** (a) Diabatic trans population of the 25D retinal model. (b) Diabatic ground state population of the 25D retinal model. rma is the maximally allowed TT-rank, and the reported values are determined by gradual increasing the rank until population curves converge.
Figure 6 shows a detailed comparison of the time-dependent reduced probability densities obtained with TT-SOKSL and TT-KSL as a function of the two large amplitude coordinates $\theta$ and $q_c$, after integrating out the bath degrees of freedom. The results show that the wavepacket dynamics is essentially identical for both methods, as shown by the reduced probability density, $\rho(t; \theta, q_c) = \int dz \Psi(t; \theta, q_c, z)^* \Psi(t; \theta, q_c, z)$ in the $S_0$ and $S_1$ electronic states.

Figure 6: Reduced probability density, $\rho(t; \theta, q_c) = \int dz \Psi(t; \theta, q_c, z)^* \Psi(t; \theta, q_c, z)$, over the two primary reactive coordinates, where $z = q_1, ..., q_{23}$. The cis regime is shaded.

Clearly, the probability densities produced by the two methods agree very closely. It is worth noting that, for this particular model system, the $S_1$ wavepacket reaches the edge of the simulation box at 150-200 fs, which causes strong oscillations due to non-adiabatic dynamics. Up to that time, the wavepacket dynamics simulated with TT-SOKSL and TT-KSL agree very well with TT-SOFT, which suggests that small discrepancies at longer times arise due to the dependency of the periodic model potential on the isomerization dihedral angle, $\theta$.  

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Figure 7 shows the calculated electronic absorption spectrum obtained by TT-SOKSL and TT-SOFT, and ML-MCTDH result, and shows that the three methods generate a nearly identical electronic absorption spectrum for the 25D retinal model, which further illustrates the capabilities of TT-SOKSL as compared to other state-of-the-art methods.

Figure 7: Electronic absorption spectrum. ML-MCTDH result reproduced from ref. 74

Figure 8 shows the analysis of the time-dependent TT-ranks for the simulation of the 25-dimensional model system. As for the two-dimensional model, TT-SOKSL enables lower-rank representation than TT-SOFT. During the first 100 fs, the growth of TT-rank is relatively fast for TT-SOFT, reaching 52 at 100 fs, whereas TT-SOKSL requires only a TT-rank of 16, very similar to TT-KSL.

Figure 8: TT-rank of the wavefunction for the 25D retinal model at short time.

Figure 9 shows the analysis of norm conservation. Clearly, TT-SOKSL agrees well with
the level of norm conservation of TT-KSL, whereas TT-SOFT exhibits a significant loss as the propagation proceeds beyond the time when the wavepacket reaches the edge of the simulation box, which suggests superior accuracy of TT-SOKSL for systems with high complexity.

![Figure 9: Norm conservation for the 25D retinal model.](image)

### 4 Discussion and Concluding Remarks

We have introduced the TT-SOKSL method for quantum simulations of multidimensional model systems. We have demonstrated the capabilities of TT-SOKSL as applied to simulations of non-adiabatic quantum dynamics for a two-state 25-dimensional model system corresponding the photoisomerization of the retinyl chromophore in rhodopsin. TT-SOKSL propagates the quantum state represented as a tensor train using the Trotter expansion of the time-evolution operator as in TT-SOFT. However, the exponential operators of the Trotter expansion are implemented in TT-SOKSL using the KSL algorithm. Therefore, TT-SOKSL exploits the benefits of both TT-SOFT and TT-KSL methods. Like TT-SOFT, it avoids the need to implement matrix-product operators (e.g., the Laplacian operator of the kinetic energy) by transforming the TT representation to momentum space and exploits the linear scaling of multidimensional TT Fourier transforms. Like TT-KSL, it exploits the advantages of projector splitting for efficiently evolving the quantum state on low-rank
We have demonstrated that TT-SOKSL provides two computational advantages over TT-SOFT as applied to simulations of multidimensional quantum dynamics. First, TT-SOKSL allows for more efficient (low-rank) tensor train representations when compared to the propagation scheme based on scaling and squaring followed by rounding in TT-SOFT. Second, TT-SOKSL allows for better norm conservation when applied with limited rank, even when the complexity of the time-dependent state increases (e.g., due to non-adiabatic effects and interferences in a periodic potential). Nevertheless, when applied with unlimited maximum rank, both TT-SOKSL and TT-SOFT conserve the norm since they are both formally unitary.150 So, differences in norm conservation when comparing TT-SOKSL, TT-KSL, and TT-SOFT result from the slightly different performance of projection versus rounding to reach a lower-rank manifold. In TT-SOFT, the wavepacket is rounded after each propagation step according to a maximum TT-rank or desired precision.

However, the truncation can compromise normalization. On the other hand, the KSL algorithm enforces a fixed rank and the projection onto the tangent space ensures a minimal distance to the true solution. Therefore, no truncation is necessary in TT-KSL and TT-SOKSL and the propagation makes optimal utilization of the low-rank subspace.

We have shown how the TT-SOKSL method avoids the need of a matrix product operator, representing the kinetic energy as a tensor train in momentum space. In contrast to the TT-KSL method that requires a matrix Hamiltonian based on a finite difference or Fourier Grid Hamiltonian such as the DVR, TT-SOKSL operates with diagonal (vector tensor trains) operators. Therefore, TT-SOKSL has the advantage of reduced memory requirement and vector-vector multiplications when compared to the matrix-vector multiplication of TT-KSL.

Finally, we note that the strategy of TT-SOKSL—combining the split-operator Hamiltonian and the efficient KSL projection scheme—could be exploited in other quantum propagation methods. For example, in Chebyshev propagation, the Hamiltonian is represented via the Chebyshev expansion, and the Chebyshev polynomials can be represented as a tensor.
So, the TT-SOKSL splitting could provide a more effective scheme to reduce the TT-rank of the Hamiltonian. Solving the split equation with the TT-KSL scheme could provide further computational advantage such as norm conservation and efficient utilization of a low-rank tensor-train array. Therefore, we anticipate the strategy of TT-SOKSL can help facilitate the development of quantum dynamical methods for a wide range of applications.

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A Dynamical Low-Rank Approximation Method

The dynamical low-rank approximation method provides an approximate rank $r$ solution to the differential equation,

$$\dot{\Psi}(t) = M(\Psi(t)).$$

(22)

The method can be applied for wavepacket propagation when $\Psi(t)$ is the time-dependent wavepacket describing the evolution of the system, since Eq. (22) is the time-dependent Schrödinger equation when $M = -i\hat{H}/\hbar$ is defined in terms of the Hamiltonian $\hat{H}$. In TT-SOKSL, however, the dynamical low-rank approximation is implemented, according to Eq. (12), for each of the terms of the Trotter factorization. Therefore, $M$ is defined by either the kinetic, or the potential energy term of the Hamiltonian.

Initially, the wavepacket is reshaped as a matrix $Y \in \mathbb{C}^{m \times n}$ and decomposed as a rank-$r$
product of three full-rank matrices (e.g., by QR decomposition), as follows:

$$Y(t) = U(t)S(t)V(t)^\dagger,$$  \hspace{1cm} (23)

where the dagger denotes the adjoint matrix (i.e., conjugate transpose), $U(t) \in \mathbb{C}^{m \times r}$ and $V(t) \in \mathbb{C}^{n \times r}$ have $r$ orthonormal columns, and $S(t) \in \mathbb{C}^{r \times r}$ is invertible (not necessarily diagonal).

We ensure that $Y(t)$ evolves on the manifold of rank $r$ by propagating the matrices $U(t)$, $S(t)$, and $V(t)$ subject to the orthonormality conditions of Stiefel manifolds, $U(t)^\dagger U(t) = I$ and $V(t)^\dagger V(t) = I$. Therefore,

$$\dot{U}(t)^\dagger U(t) + c.c. = 0,$$
$$\dot{V}(t)^\dagger V(t) + c.c. = 0,$$

where c.c. denotes the complex conjugate of the preceding terms. So, the evolution preserves the number $r$ of linearly independent columns of $U(t)$ and $V(t)$, which in turn preserves the rank of $Y(t)$, as shown below with the discussion of Eq. (29).

There are many ways of enforcing Eq. (24) and therefore many possible solutions. Nevertheless, a unique solution is obtained by adopting the following gauge conditions:

$$\dot{U}(t)^\dagger U(t) = 0,$$
$$\dot{V}(t)^\dagger V(t) = 0,$$  \hspace{1cm} (25)

leading to unique equations of motion for $U(t)$, $V(t)$, and $S(t)$, as shown in Appendix B:

$$\dot{S}(t) = U^\dagger M(Y)V,$$
$$\dot{U}(t) = (1 - UU^\dagger)M(Y)VS^{-1},$$
$$\dot{V}(t) = (1 - VV^\dagger)M(Y)^\dagger US^{-\dagger}.$$  \hspace{1cm} (26)
Integrating the equations of motion introduced by Eq. (26) by using standard numerical techniques (e.g., Runge-Kutta, Velocity-Verlet) becomes challenging when $S$ has very small singular values since $S^{-1}$ becomes an ill-conditioned matrix with large norm, so a very small integration time-step is required. In fact, this is what usually happens when the actual rank of the exact solution is smaller than $r$, as in calculations where the rank is not known and thus overestimated to ensure an accurate approximation. A similar problem arises when integrating the equations of motion of the MCTDH method since they depend on the inverse of the density matrix. That problem is usually addressed by some sort of regularization scheme, although regularization introduces errors that are uncertain. In contrast, the dynamical low-rank approximation method bypasses the need to integrate equations with singular matrices (i.e., Eq. (26)) simply by directly integrating the equation of motion of $Y$ – i.e., Eq. (29), where there is no singular matrix since $S^{-1}$ cancels with $S$.

The equation of motion for $Y$ is obtained from Eq. (23), as follows:

$$\dot{Y} = \dot{U}SV^\dagger + U\dot{S}V^\dagger + US\dot{V}^\dagger,$$

(27)

and, substituting $\dot{U}$, $\dot{S}$, and $\dot{V}$ according to Eq. (26), we obtain:

$$\dot{Y} = (1 -UU^\dagger)M(Y)VS^{-1}SV^\dagger + UU^\dagger M(Y)V V^\dagger + US((1 -VV^\dagger)M(Y)^\dagger US^{-\dagger})^\dagger,$$

(28)

which gives the equation of motion of $Y$, as follows:

$$\dot{Y} = M(Y)VV^\dagger - UU^\dagger M(Y)VV^\dagger + UU^\dagger M(Y),$$

(29)

Equation (29) is efficiently integrated by using the Strang splitting approximation, as outlined in Appendix D. Note that each of the terms on the right-hand-side (RHS) of Eq. (29) involves a projection operator $P_U = UU^\dagger$, or $P_V = VV^\dagger$, which ensures that $\dot{Y}$ does not have any component orthogonal to the manifold of rank $r$. The first term corresponds to
evolution of \( U \) and \( S \) at constant \( V \). The second term evolves \( S \) at constant \( U \) and \( V \), and the third term evolves \( S \) and \( V \) at constant \( U \).

### A.1 Projection onto the tangent plane

Comparing Eq. (29) and Eq. (13), we can readily identify \( P_Y \), as follows:

\[
\dot{Y} = P_Y(M(Y)) = M(Y)VV^\dagger - UU^\dagger M(Y)VV^\dagger + UU^\dagger M(Y).
\]

Therefore, the projection of any arbitrary state \( Z \) onto the ‘tangent plane’ (i.e., the vector space tangent to the manifold of rank \( r \) at \( Y \)) can be defined, as follows:

\[
P_Y(Z) = ZVV^\dagger - UU^\dagger ZVV^\dagger + UU^\dagger Z,
\]

where the three terms on the RHS of Eq. (31) involve the projection operator \( P_U = UU^\dagger \), or \( P_V = VV^\dagger \), so they are on the tangent plane and thus invariant under the effect of \( P_Y \). As an example, we show that \( P_Y \) does not change the middle term of Eq. (31), as follows:

\[
P_Y(-UU^\dagger ZVV^\dagger)
\]

\[
= (-UU^\dagger ZVV^\dagger)VV^\dagger - UU^\dagger (-UU^\dagger ZVV^\dagger)VV^\dagger + UU^\dagger (-UU^\dagger ZVV^\dagger)
\]

\[
= -UU^\dagger ZVV^\dagger + UU^\dagger ZVV^\dagger - UU^\dagger ZVV^\dagger
\]

\[
= -UU^\dagger ZVV^\dagger.
\]

Therefore, evolving a rank-\( r \) state \( Y \) by displacement along the direction of \( \dot{Y} \), as defined in Eq. (29), always generates a rank-\( r \) state regardless of the rank of \( M(Y) \). The resulting propagation avoids the need to first generate a high-rank state \( M(Y) \) and then reduce its rank by singular value decomposition or by projection onto the tangent manifold, since \( \dot{Y} \) generates a displacement on the tangent plane.
B  Equations of motion for U, S and V

We obtain the equations of motion introduced by Eq. (26) by taking the time derivative of Eq. (23), as follows:

\[ \dot{Y} = \dot{U}SV^\dagger + U\dot{S}V^\dagger + US\dot{V}^\dagger, \]  

(33)

and imposing the gauge conditions, as follows.

B.1  Equation of motion for S

We invoke orthonormality \( V^\dagger V = U^\dagger U = I \) with the gauge conditions (i.e., \( U^\dagger \dot{U} = 0 \) and \( V^\dagger \dot{V} = 0 \), implying \( (V^\dagger \dot{V})^\dagger = \dot{V}^\dagger V = 0 \)) to obtain:

\[
U^\dagger \dot{Y}V = U^\dagger \dot{U}SV^\dagger V + U^\dagger US\dot{V}^\dagger V + U^\dagger USV^\dagger \dot{V}, 
\]

(34)

Next, we show below that

\[ U^\dagger \dot{Y}V = U^\dagger M(Y) V, \]  

(35)

and, substituting Eq. (35) into Eq. (34), we obtain the equation of motion for S, as introduced in Eq. (26):

\[ \dot{S}(t) = U^\dagger M(Y) V. \]  

(36)

Equation (35) is obtained by first noting that the error \( \epsilon = M(Y) - \dot{Y} \) must be orthogonal to any state \( \delta Y \) in the tangent plane –i.e. \( \langle \dot{Y} - M(Y) | \delta Y \rangle = 0 \). In particular, for the tangent vectors \( \delta Y = U_i V_j^\dagger \) with \( i, j = 1, \ldots, r \) (corresponding to vectors of the form \( \delta Y = \delta U \delta SV^\dagger + \delta USV^\dagger + US\delta V^\dagger \), with \( U_i \) the \( i^{th} \) column of \( U \), \( V_j \) the \( j^{th} \) column of \( V \), \( \delta U = \delta V = 0 \), and \( \delta S_{kl} = \delta_{ki}\delta_{lj} \), we obtain: \( \langle U_i V_j^\dagger | \dot{Y} - M(Y) \rangle = 0 \). Rearranging that
inner product, we obtain:

\[ \langle U_i V_j^\dagger | \dot{Y} \rangle = \langle U_i V_j^\dagger | M(Y) \rangle. \quad (37) \]

Next, we note that \( \langle U_i V_j^\dagger | Z \rangle = U_i^\dagger Z V_j \) where \( Z = \{ \dot{Y}, M(Y) \} \) since

\[
\langle U_i V_j^\dagger | Z \rangle = \sum_{k,l}^{m,n} (U_i V_j^\dagger)_{k,l} Z_{k,l} \\
= \sum_k^m \sum_l^n U_{ki}^* V_{jl} Z_{kl} \\
= \sum_k^m \sum_l^n U_{ki}^* Z_{kl} V_{jl} \\
= U_{1i}^* \sum_l^n Z_{1l} V_{jl} + \ldots + U_{mi}^* \sum_l^n Z_{ml} V_{jl} \\
= U_i^\dagger \begin{pmatrix} \sum_l^n Z_{1l} V_{jl} \\ \vdots \\ \sum_l^n Z_{ml} V_{jl} \end{pmatrix} \\
= U_i^\dagger Z V_j. \quad (38)
\]

Substituting Eq. (38) into Eq. (37), we obtain:

\[ U_i^\dagger \dot{Y} V_j = U_i^\dagger M(Y) V_j, \quad (39) \]

which proves Eq. (35).

### B.2 Equation of motion for U

We obtain the equation of motion for \( U \), as introduced in Eq. (26), by choosing the tangent vector \( \delta Y = \delta u (\sum_j^r S_j V_j^\dagger) \), where \( \delta u \) is a length-\( m \) vector that satisfies the condition \( U_i^\dagger \delta u = 0 \), where \( V_j \) is the \( j^{th} \) column of \( V \). With this \( \delta Y \), we have the orthogonality condition
\[ \langle M(Y) - \dot{Y}|\delta u(\sum_j^r S_{ij} V_j^\dagger)\rangle = 0, \] and rearranging the inner product, we obtain:

\[ \langle \delta u(\sum_j^r S_{ij} V_j^\dagger)|\dot{Y}\rangle = \langle \delta u(\sum_j^r S_{ij} V_j^\dagger)|M(Y)\rangle. \tag{40} \]

Using the equivalence between the inner product and vector-matrix-vector product, introduced by Eq. (38), the two sides of Eq. (40) can be written, as follows:

\[ \delta u^\dagger \dot{Y} \sum_j^r S_{ij} V_j = \delta u^\dagger M(Y) \sum_j^r S_{ij} V_j, \tag{41} \]

and introducing the substitution \( \sum_j^r S_{ij} V_j = VS_i \) into Eq. (41), we obtain:

\[ \delta u^\dagger \dot{Y} VS_i = \delta u^\dagger M(Y) VS_i. \tag{42} \]

Using the condition that \( S \) is invertible, we obtain:

\[ \delta u^\dagger \dot{Y} V = \delta u^\dagger M(Y) V, \tag{43} \]

and substituting \( \dot{Y} \) in Eq. (43), according to Eq. (27), we obtain:

\[ \delta u^\dagger (USV^\dagger + US^\dagger V + USV^\dagger) V = \delta u^\dagger M(Y) V, \tag{44} \]

that can be further simplified using \( \delta u^\dagger U = 0 \), as follows:

\[ \delta u^\dagger USV^\dagger V = \delta u^\dagger M(Y) V. \tag{45} \]

Further, considering that \( V^\dagger V = I \) and \( S \) is invertible, we obtain:

\[ \delta u^\dagger \dot{U} = \delta u^\dagger M(Y) VS^{-1}, \tag{46} \]
so,
\[ \delta u^\dagger (\dot{U} - M(Y)VS^{-1}) = 0. \]  
(47)

Considering that Eq. (47) is satisfied by all \( \delta u \), so long as \( \delta u^\dagger U = 0 \), Eq. (47) implies that \( \dot{U} - M(Y)VS^{-1} = 0 \) or \( \dot{U} - M(Y)VS^{-1} = \lambda U \) where \( \lambda \) is a number, so

\[ (I - UU^\dagger)(\dot{U} - M(Y)VS^{-1}) = 0, \]  
(48)

where the effect of the projector \((I - UU^\dagger)\) is to enforce the gauge condition \( \delta u^\dagger U = 0 \). Rearranging Eq. (48), we obtain:

\[ (I - UU^\dagger)\dot{U} = (I - UU^\dagger)(M(Y)VS^{-1}), \]  
(49)

and considering that \( U^\dagger \dot{U} = 0 \), we obtain the equation of motion for \( U \), as follows:

\[ \dot{U} = (I - UU^\dagger)(M(Y)VS^{-1}). \]  
(50)

**B.3 Equation of motion for V**

The equation of motion for \( V \) is obtained analogously, using the tangent vector \( \delta Y = (\sum_r U_j S_{ji}) \delta v^\dagger \), with \( \delta v \) a length-\( n \) vector that satisfies the condition \( V^\dagger \delta v = 0 \), which must fulfill the orthogonality condition \( \langle \delta Y | \dot{Y} - M(Y) \rangle = 0 \):

\[ \langle \sum_j^r U_j S_{ji} \delta v^\dagger | \dot{Y} \rangle = \langle \sum_j^r U_j S_{ji} \delta v^\dagger | M(Y) \rangle, \]  
(51)

which can be written as a vector-matrix-vector product, similar to Eq. (41):

\[ (\sum_j^r U_j^\dagger S_{ji}) \dot{Y} \delta v = (\sum_j^r U_j^\dagger S_{ji})M(Y)\delta v. \]  
(52)
Substituting $\dot{Y}$ into Eq. (52), according to Eq. (27), we obtain:

\[
(\sum_j U_j^\dagger S_{ji})(\dot{U}SV^\dagger + USV^\dagger + US\dot{V}^\dagger)\delta v = (\sum_j U_j^\dagger S_{ji})M(Y)\delta v, \quad (53)
\]

which is simplified with $V^\dagger \delta v = 0$, as follows:

\[
(\sum_j U_j^\dagger S_{ji})US\dot{V}^\dagger = (\sum_j U_j^\dagger S_{ji})M(Y)\delta v. \quad (54)
\]

Next, we take the complex transpose (adjoint) of Eq. (54) to obtain an explicit expression of $\dot{V}$, as follows:

\[
\delta v^\dagger \dot{V}S^\dagger U^\dagger (\sum_j U_j S_{ji}) = \delta v^\dagger (M(Y))^\dagger (\sum_j U_j S_{ji}). \quad (55)
\]

and substituting the matrix-vector product, $\sum_j U_j S_{ji} = US_i$, we obtain:

\[
\delta v^\dagger \dot{V}S^\dagger U^\dagger US_i = \delta v^\dagger (M(Y))^\dagger US_i, \quad (56)
\]

which is further simplified using $U^\dagger U = I$:

\[
\delta v^\dagger \dot{V}S^\dagger S_i = \delta v^\dagger (M(Y))^\dagger US_i. \quad (57)
\]

As before, we multiply both sides with $S_i^{-1}$ to obtain:

\[
\delta v^\dagger \dot{V}S^\dagger = \delta v^\dagger (M(Y))^\dagger U, \quad (58)
\]

and multiplying both sides by the inverse of $S^\dagger$ ($i.e., S^{-\dagger}$), we obtain:

\[
\delta v^\dagger (\dot{V} - (M(Y))^\dagger US^{-\dagger}) = 0, \quad (59)
\]
implying that $\dot{V} - (M(Y))^\dagger US^{-\dagger}$ is orthogonal to $\delta v^\dagger$, or equal to zero. Therefore,

$$(I - VV^\dagger)(\dot{V} - (M(Y))^\dagger US^{-\dagger}) = 0. \tag{60}$$

Finally, considering the gauge condition $V^\dagger \dot{V} = 0$, we obtain:

$$\dot{V} = (I - VV^\dagger)(M(Y))^\dagger US^{-\dagger}. \tag{61}$$

**B.4 Mutually orthogonal subsets**

The tangent vectors chosen to obtain the equations for $S$, $U$ and $V$ define the subsets,

$$V_S = \{USV^\dagger, USV^\dagger U^\dagger \delta U = 0\},$$

$$V_U = \{\delta USV^\dagger, U^\dagger \delta U = 0\},$$

$$V_V = \{US \delta V^\dagger, V^\dagger \delta V = 0\}, \tag{62}$$

which are mutually orthogonal since the inner product between elements from any pair of subsets is equal to zero. For example, $\langle U^\dagger \delta SV^\dagger | \delta USV^\dagger \rangle = \text{Tr}[V^\dagger \delta S^\dagger U^\dagger \delta USV^\dagger] = 0$ since $U^\dagger \delta U = 0$. Analogously, $\langle U^\dagger \delta SV^\dagger | US^\dagger \delta V^\dagger \rangle = \text{Tr}[V^\dagger \delta S^\dagger U^\dagger US^\dagger \delta V^\dagger] = \text{Tr}[V^\dagger \delta V^\dagger V^\dagger \delta S^\dagger U^\dagger US^\dagger] = 0$ since $\delta V^\dagger V = 0$. Finally, $\langle \delta USV^\dagger | US^\dagger \delta V^\dagger \rangle = \text{Tr}[V^\dagger \delta S^\dagger U^\dagger US^\dagger \delta V^\dagger] = 0$ since $\delta U^\dagger U = 0$.

Together, the three subsets define the complete set of tangent vectors $T_Y M_r^{m \times n}$, according to the following direct sum of mutually orthogonal subsets:

$$T_Y M_r^{m \times n} = V_U \oplus V_S \oplus V_V, \tag{63}$$

$$= \{\delta Y = \delta USV^\dagger + U^\dagger \delta SV^\dagger + US \delta V^\dagger, U^\dagger \delta U = 0, V^\dagger \delta V = 0\},$$

where the curly bracket denotes the set of tangent vectors, with $\delta S \in \mathbb{C}^{r \times r}$, $\delta U \in \mathbb{C}^{m \times r}$, and $\delta V \in \mathbb{C}^{n \times r}$ fulfilling the gauge conditions.

The mutually orthogonal relationship unifies the derivation of the equations of motion.
for $U$, $S$, and $V$, as follows:

$$\langle M(Y) - \dot{Y} | \delta Y \rangle = 0 \text{ for any } \delta Y \in T_Y \mathcal{M}. \quad (64)$$

When $\delta Y = \delta USV^\dagger$,

$$\langle M(Y) - \dot{Y} | \delta USV^\dagger \rangle = \langle M(Y) - \dot{USV}^\dagger - \dot{USV}^\dagger | \delta USV^\dagger \rangle = 0, \quad (65)$$

and considering that $U^\dagger \delta U = 0$, $\langle U\dot{SV}^\dagger - US\dot{V}^\dagger | \delta USV^\dagger \rangle = \langle U(\dot{SV}^\dagger - \dot{S}V^\dagger) | \delta USV^\dagger \rangle = \text{Tr}[(\dot{VS}^\dagger - \dot{V}S^\dagger)U^\dagger \delta USV^\dagger] = 0$, we obtain:

$$\langle M(Y) - \dot{Y} | \delta USV^\dagger \rangle = \langle M(Y) - \dot{USV}^\dagger | \delta USV^\dagger \rangle = 0. \quad (66)$$

Rearranging Eq. (66), we obtain:

$$\langle M(Y) - \dot{USV}^\dagger | \delta USV^\dagger \rangle = \langle M(Y) | \delta USV^\dagger \rangle - \langle \dot{USV}^\dagger | \delta USV^\dagger \rangle$$

$$= \text{Tr}[M(Y)^\dagger \delta USV^\dagger] - \text{Tr}[VS^\dagger \dot{U}^\dagger \delta USV^\dagger]$$

$$= \text{Tr}[SV^\dagger M(Y)^\dagger \delta U] - \text{Tr}[SV^\dagger VS^\dagger \dot{U}^\dagger \delta U]$$

$$= \text{Tr}[SV^\dagger M(Y)^\dagger \delta U] - \text{Tr}[SS^\dagger \dot{U}^\dagger \delta U]$$

$$= \langle M(Y) VS^\dagger | \delta U \rangle - \langle \dot{US}^\dagger SS^\dagger | \delta U \rangle$$

$$= \langle M(Y) VS^\dagger - \dot{US}^\dagger SS^\dagger | \delta U \rangle, \quad (67)$$

so, according to Eqs. (66) and (67), $\langle M(Y) VS^\dagger - \dot{US}^\dagger SS^\dagger | \delta U \rangle = 0$, which implies that $M(Y) VS^\dagger - \dot{US}^\dagger SS^\dagger$ is orthogonal to $\delta U$ (i.e., parallel to $U$ since $U^\dagger \delta U = 0$). So,

$$\langle I - UU^\dagger | M(Y) VS^\dagger - \dot{US}^\dagger SS^\dagger \rangle = \langle I - UU^\dagger | M(Y) VS^\dagger - \dot{US}^\dagger SS^\dagger \rangle = 0, \quad (68)$$

where we used the gauge condition $U^\dagger \dot{U} = 0$. Rearranging the second equality of Eq. (68)
leads to the equation of motion for $U$:

$$
\dot{U}SS^\dagger = (I -UU^\dagger)M(Y)VS^\dagger, \quad (69)
$$

$$
\dot{U} = (I -UU^\dagger)M(Y)VS^{-1}.
$$

Analogously, choosing $\delta Y = U\delta SV^\dagger$ in Eq. (64) leads to:

$$
\langle M(Y) - U\dot{SV}^\dagger | U\delta SV^\dagger \rangle = 0, \quad (70)
$$

which, with a procedure similar to Eq. (67), leads to:

$$
\langle \dot{S} | \delta S \rangle = \langle U^\dagger M(Y)V | \delta S \rangle. \quad (71)
$$

Therefore,

$$
\dot{S} = U^\dagger M(Y)V. \quad (72)
$$

As for the derivation of $\dot{U}$, choosing $\delta Y = US\delta V^\dagger$ in Eq. (64) gives the equation for $V$:

$$
\dot{V} = (I -VV^\dagger)M(Y)^\dagger US^{-\dagger}. \quad (73)
$$

## C KSL Integration

The equation of motion for $Y$, introduced by Eq. (30),

$$
\dot{Y} = M(Y)VV^\dagger - UUU^\dagger M(Y)VV^\dagger + UUU^\dagger M(Y), \quad (74)
$$
is integrated by sequentially applying the three terms on the RHS of Eq. (74) according to the following Lie-Trotter splitting method over the time interval $[t_0, t_1]$:

$$
\dot{Y}_I = M(Y_0)V_I V_I^\dagger, \quad \text{with } Y_I(t_0) = U_I S_I V_I^\dagger = Y_0
$$

$$
\dot{Y}_{II} = -U_{II} U_{II}^\dagger M(Y_0)V_{II} V_{II}^\dagger, \quad \text{with } Y_{II}(t_0) = U_{II} S_{II} V_{II}^\dagger = Y_I(t_1)
$$

$$
\dot{Y}_{III} = U_{III} U_{III}^\dagger M(Y_0), \quad \text{with } Y_{III}(t_0) = U_{III} S_{III} V_{III}^\dagger(t_0) = Y_{II}(t_1)
$$

which gives the approximation $Y(t_1) \approx Y_{III}(t_1)$. Note that the first equation of motion, introduced by the splitting scheme of Eq. (75), evolves $Y = USV^\dagger$ at constant $V$ (i.e., $\dot{V}_I = 0$) due to the effect of projection operator $P_{V_I} = V_I V_I^\dagger$, which effectively propagates only the product $K = US$. Considering that $\dot{Y}_I = M(Y_0)V_I V_I^\dagger = \dot{K}V_I^\dagger$, we obtain $\dot{K} = M(Y_0)V_I$. The second equation of Eq. (75) evolves $S$ at constant $U$ and $V$, and the third equation evolves the product $L = SV^\dagger$ at constant $U$. Therefore, the resulting method avoids equations of motion with singular matrices by sequentially propagating the matrices $K$, $S$, and $L$ (i.e., by KSL propagation).

C.1 Integration of $K$

We solve the first equation of motion, introduced by Eq. (75),

$$
\dot{Y}_I = M(Y_0)V_I V_I^\dagger,
$$

using the initial condition, $Y_I(t_0) = Y_0$ with $Y_I = U_I S_I V_I^\dagger$, and $\dot{V}_I = 0$, so

$$
\dot{Y}_I = \dot{K}V_I^\dagger,
$$

(77)
with $K = U_I S_I$. Integrating Eq. (77) with the trapezoidal rule, we obtain:

$$
\int_{t_0}^{t_1} dt \dot{Y}_I = \int_{t_0}^{t_1} dt K V_I^\dagger,
$$

(78)

$$
Y_I(t_1) - Y_I(t_0) \approx (t_1 - t_0) M(Y_0) V_I(t_0) V_I^\dagger(t_0).
$$

Therefore,

$$
Y_I(t_1) \approx (U_I(t_0) S_I(t_0) + (t_1 - t_0) M(Y_0) V_I(t_0)) V_I^\dagger(t_0).
$$

(79)

C.2 Integration of $S$

Having obtained $Y_I(t_1) = U_I(t_1) S_I(t_1) V_I(t_0)^\dagger$, we compute $U_I(t_1)$ and $S_I(t_1)$ by QR decomposition of $K_I(t_1) = Y_I(t_1) V_I(t_0)$ (i.e., $QR[K_I(t_1)] \rightarrow U_I(t_1), S_I(t_1)$), and we initialize $Y_{II} = U_{II} S_{II} V_{II}^\dagger$ with $U_{II}(t_0) = U_I(t_1)$, $S_{II}(t_0) = S_I(t_1)$ and $V_{II}(t_0) = V_I(t_0)$. We integrate the equation:

$$
\dot{Y}_{II} = -U_{II} U_{II}^\dagger M(Y_0) V_{II}^\dagger,
$$

(80)

keeping constant $U_{II}$ and $V_{II}$, and effectively integrate the equation $\dot{S}_{II} = -U_{II}^\dagger M(Y_0) V_{II}$ from $t_0$ to $t_1$ with the trapezoidal rule, as follows:

$$
S_{II}(t_1) \approx S_{II}(t_0) - U_{II}^\dagger(t_0)(t_1 - t_0) M(Y_0) V_{II}(t_0),
$$

(81)

to obtain $Y_{II}(t_1)$, as follows:

$$
Y_{II}(t_1) = U_{II}(t_0) S_{II}(t_1) V_{II}^\dagger(t_0).
$$

(82)

C.3 Integration of $L$

Finally, we initialize $Y_{III} = U_{III} S_{III} V_{III}^\dagger$ with $U_{III}(t_0) = U_{II}(t_0)$, $S_{III}(t_0) = S_{II}(t_1)$ and $V_{III}(t_0) = V_{II}(t_0)$, and we solve the equation $\dot{Y}_{III} = U_{III} U_{III}^\dagger M(Y_0)$, with constant $U_{III}$,
which effectively propagates only the product $L = S_{III} V_{III}^\dagger$. Considering that
\[
\dot{Y}_{III} = U_{III} \dot{L},
\] (83)
we obtain $\dot{L} = U_{III}^\dagger M(Y_0)$, which upon integration by the trapezoidal rule gives
\[
L(t_1) \approx L(t_0) + U_{III}(t_0)^\dagger M(Y_0)(t_1 - t_0),
\] (84)
with $S_{III}(t_1)$ and $V_{III}(t_1)$ obtained by QR decomposition of $L(t_1)$.

Integrating Eq. (83), we obtain:
\[
Y_{III}(t_1) - Y_{III}(t_0) = U_{III}(t_0)(L(t_1) - L(t_0)),
\] (85)
so, $Y_{III}(t_1) = U_{III}(t_0)L(t_1) = U_{III}(t_0)S_{III}(t_1)V_{III}(t_1)^\dagger$. The approximation $Y(t_1) \approx Y_{III}(t_1)$ completes the propagation step from $t_0$ to $t_1$. The next integration step is initialized, as follows: $U_0 = U_{III}(t_1)$, $S_0 = S_{III}(t_1)$, and $V_0 = V_{III}(t_1)$, such that $Y_0 = Y(t_1)$.

\section{TT notation}

This Appendix introduces the TT notation necessary for the derivation of the TT-KSL equations of motion, including left and right unfoldings and orthogonalizations, partial products, and reconstructions, consistent with the TT literature.\cite{3661173}

\subsection{Tensor unfolding and reconstruction}

The $i$-th unfolding of a tensor $X \in \mathbb{C}^{n_1 \times \cdots \times n_d}$ involves reshaping the tensor as a matrix $X^{(i)} \in \mathbb{C}^{(n_1 n_2 \cdots n_i) \times (n_{i+1} \cdots \times n_d)}$, with entries $X_{jk}^{(i)}$ corresponding to row $j = n_1 n_2 \cdots n_i$ and column $k = n_{i+1} \cdots \times n_d$. Tensor reconstruction is the inverse of unfolding, indicated as
follows:

$$X = \text{Ten}_i[X^{(i)}].$$  \hfill (86)

### D.2 TT core unfoldings

TT cores $X_i \in \mathbb{C}^{r_i-1 \times n_i \times r_i}$ can be matricized in terms of the so-called left or right unfoldings, denoted as $X_i^\leftarrow \in \mathbb{C}^{r_i-1 \times n_i \times r_i}$ and $X_i^\rightarrow \in \mathbb{C}^{r_i \times n_i \times r_i-1}$, respectively, as follows:

$$X_i^\leftarrow = \left( \begin{array}{c}
X_i(:, 1, :)
\vdots
\vdots
X_i(r_i-1, 1, 1) & \cdots & X_i(r_i-1, 1, r_i)
\vdots
X_i(1, 2, 1) & \cdots & X_i(1, 2, r_i)
\vdots
\vdots
X_i(r_i-1, 2, 1) & \cdots & X_i(r_i-1, 2, r_i)
\vdots
\vdots
X_i(1, n_i, 1) & \cdots & X_i(1, n_i, r_i)
\vdots
\vdots
X_i(:, n_i, :)
\end{array} \right) = \left( \begin{array}{c}
X_i(1, 1, 1) & \cdots & X_i(1, 1, r_i)
\vdots
\vdots
X_i(r_i-1, 1, 1) & \cdots & X_i(r_i-1, 1, r_i)
\vdots
X_i(1, 2, 1) & \cdots & X_i(1, 2, r_i)
\vdots
\vdots
X_i(r_i-1, 2, 1) & \cdots & X_i(r_i-1, 2, r_i)
\vdots
\vdots
X_i(1, n_i, 1) & \cdots & X_i(1, n_i, r_i)
\vdots
\vdots
X_i(:, n_i, :)
\end{array} \right)$$  \hfill (87)
D.3 Partial products of TT cores

The left partial product \( X_{\leq i} \in \mathbb{C}^{n_1 \times \ldots \times n_i \times r_i} \) is defined, as follows:

\[
X_{\leq i}(l_1, \ldots, l_i, a_i) = X_1(l_1) \cdots X_i(l_i), \tag{89}
\]

where \( a_i = 1, \ldots, r_i \) and \( l_k = 1, \ldots, n_k \) for \( k = 1, \ldots, i \). Analogously, the right partial product \( X_{\geq i+1} \in \mathbb{C}^{n_i \times n_{i+1} \times \ldots \times n_d} \) is defined, as follows:

\[
X_{\geq i+1}(a_i, l_{i+1}, \ldots, l_d) = X_{i+1}(l_{i+1}) \cdots X_d(l_d). \tag{90}
\]

Therefore, the unfoldings \( X_{\leq i} \in \mathbb{C}^{n_1 \ldots n_i} \times r_i \) and \( X_{\geq i+1} \in \mathbb{C}^{n_{i+1} \ldots n_d} \times r_i \) of the left and right partial products, introduced by Eqs. \((89)\) and \((90)\), define the \( i \)-th unfolding of the tensor, as follows:

\[
X^{(i)} = X_{\leq i} X_{\geq i+1}^\dagger, \tag{91}
\]

and, according to Eq. \((86)\),

\[
X = \text{Ten}_i[X_{\leq i} X_{\geq i+1}^\dagger]. \tag{92}
\]
D.4 Recursive construction

The unfoldings of left and right partial products $X_{\leq i} \in \mathbb{C}^{(n_1 \cdots n_i) \times r_i}$ and $X_{\geq i+1} \in \mathbb{C}^{(n_{i+1} \cdots n_d) \times r_i}$ can be recursively constructed, as follows:

$$X_{\leq i} = (X_{\leq i-1} \otimes I_{n_i})X^<_i,$$
$$X_{\geq i} = (I_{n_i} \otimes X_{\geq i+1})X^>_i,$$  \hspace{1cm} (93)

for $i = 1, \ldots, d$, starting with $X_{\leq 0} = X_{\geq d+1} = 1$, where $\otimes$ is the Kronecker product.

Therefore, according to Eqs. (91) and (93), we obtain:

$$X^{(i)} = (X_{\leq i-1} \otimes I_{n_i})X^<_iX^>_i,$$  \hspace{1cm} (94)

and

$$X^{(i-1)} = X_{\leq i-1}((I_{n_i} \otimes X_{\geq i+1})X^>_i)^\dagger,$$
$$= X_{\leq i-1}X^>_i((I_{n_i} \otimes X_{\geq i+1})^\dagger.$$  \hspace{1cm} (95)

D.5 Left and right orthogonalization

The recursive relations, introduced by Eq. (93), enable efficient orthogonalization of the left and right partial products by iterative QR decomposition, as follows.

Starting with $X_{\leq 2} = (X_{\leq 1} \otimes I_{n_2})X^<_2$, we perform a QR decomposition of $X_{\leq 1} = X^<_1 = Q^<_1 R_1$, with $Q_1$ and orthogonal matrix and $R_1$ an upper triangular matrix, to obtain

$$X_{\leq 2} = ((Q^<_1 R_1) \otimes I_{n_2})X^<_2,$$
$$= (Q^<_1 \otimes I_{n_2})(R_1 \otimes I_{n_2})X^<_2.$$  \hspace{1cm} (96)

Performing a QR decomposition of $(R_1 \otimes I_{n_2})X^<_2 = Q^<_2 R_2$, we obtain,

$$X_{\leq 2} = (Q^<_1 \otimes I_{n_2})Q^<_2 R_2,,$$  \hspace{1cm} (97)

39
which, according to Eq. (89), gives:

\[ X_{\leq 2} = Q_{\leq 2}R_2. \]  \hspace{1cm} (98)

Iterating \( i \) times, we obtain the left-orthogonalized partial product \( X_{\leq i} = Q_{\leq i}R_i \). Analogously, we obtain the right-orthogonalized partial product \( X_{\geq i+1} = Q_{\geq i+1}R_{i+1} \). Therefore, according to Eq. (92), we obtain:

\[ X = \text{Ten}_i[Q_{\leq i}R_iR_{i+1}^\dagger Q_{\geq i+1}^\dagger], \]  \hspace{1cm} (99)

and introducing the following substitutions \( U_{\leq i} = Q_{\leq i}, \) \( S_i = R_iR_{i+1}^\dagger, \) and \( V_{\geq i+1} = Q_{\geq i+1}, \) we obtain the SVD-like decomposition in terms of left and right orthogonalized partial products, as follows:

\[ X = \text{Ten}_i[U_{\leq i}S_iV_{\geq i+1}^\dagger]. \]  \hspace{1cm} (100)

By recursive construction (Appendix D.4), the \( i + 1 \)th unfolding matrix of a left-and-right orthogonalized \( X \) can be obtained given \( X^{(i)} = U_{\leq i}S_iV_{\geq i+1}^\dagger \). First, substituting \( V_{\geq i+1} = (V_{\geq i+2} \otimes I_{n_{i+1}})V_{\geq i+1}^\dagger \) according to Eq. (93):

\[ X^{(i)} = U_{\leq i}S_iV_{\geq i+1}^\dagger \\
= U_{\leq i}S_iV_{i+1}^\dagger(V_{\geq i+2} \otimes I_{n_{i+1}})^\dagger. \]  \hspace{1cm} (101)

Observing that Eq. (101) is Eq. (93) with \( i \) in place of \( i - 1 \), \( U_{\leq i}S_i \) in place of \( X_{\leq i-1} \), \( V_{i+1}^\dagger \) in place of \( X_{i}^\dagger \), and \( V_{\geq i+2} \) in place of \( X_{\geq i+1} \), the expression for \( X^{(i+1)} \) from Eq. (101) is
obtained by making these changes of variables in Eq. (94):

\[ X^{(i+1)} = (I_{n_i} \otimes (U_{\leq i} S_i))V_{i+1}^\leq V_{i+1}^\dagger V_{i+2}^\geq, \]  

\[ = (I_{n_i} \otimes U_{\leq i})(I_{n_i} \otimes S_i)V_{i+1}^\leq V_{i+2}^\dagger. \]

(102)

E Tangent Space and Tensor Train Projection

E.1 TT tangent space

The tangent space \( T_{Y}M_{r} \) is defined as the complete set of tensor trains that are tangent to the manifold \( M_{r} \) of tensor trains \( Y \) or rank \( r \). Analogous to the definition of the tangent space for matrices, introduced by Eq. (63), the TT tangent space is defined by the direct sum of mutually orthogonal subsets of tangent tensors, as follows:

\[ T_{Y}M_{r} = \mathcal{V}_1 \oplus \mathcal{V}_1 \oplus \cdots \oplus \mathcal{V}_d, \]  

(103)

with

\[ \mathcal{V}_j = \{ \text{Ten}_j[(U_{\leq j-1} \otimes I_{n_j})\delta C_j^\leq V_{j+1}^\dagger], \text{ where } U_j^\dagger\delta C_j^\leq = 0 \}, \text{ with } j < d, \]

\[ \mathcal{V}_d = \{ \text{Ten}_j[(U_{\leq d-1} \otimes I_{n_d})\delta C_d^\leq] \}. \]  

(104)

Note that the gauge condition \( U_j^\dagger\delta C_j^\leq = 0 \) ensures that a displacement of the tensor \( X = \text{Ten}_j[U_{\leq j} U_{j+1}^\dagger] \) along the direction of an element of \( \mathcal{V}_j \) preserves the rank since the number of linearly independent columns of \( U_j^\leq \) is preserved.

We show that the subsets \( \mathcal{V}_i \) are indeed mutually orthogonal since the inner product of any two tensors \( \delta X_j = (U_{\leq j-1} \otimes I_{n_j})\delta C_j^\leq V_{j+1}^\dagger \) and \( \delta X_k = (U_{\leq k-1} \otimes I_{n_k})\delta C_k^\leq V_{k+1}^\dagger \) from
different subsets (i.e., $V_j$ and $V_k$, with $j \neq k$) is equal to zero, as follows:

$$
\langle \delta X_j | \delta X_k \rangle = \text{Tr}[(U_{\leq j-1} \otimes I_n) \delta C_j^< V_{\geq j+1}^\dagger (U_{\leq k-1} \otimes I_n) \delta C_k^< V_{\geq k+1}^\dagger],
$$

$$
= \text{Tr}[(U_1 \cdots U_{j-1} \delta C_j^< V_{j+1}^\dagger \cdots V_d^\dagger) U_1 \cdots U_{k-1} \delta C_k^< V_{k+1}^\dagger \cdots V_d^\dagger],
$$

$$
= \text{Tr}[V_d \cdots V_{j+1} \delta C_j^< U_{j-1}^\dagger \cdots U_1^\dagger U_1 \cdots U_{k-1} \delta C_k^< V_{k+1}^\dagger \cdots V_d^\dagger],
$$

(105)

Assuming that $k > j$, and considering that the cores have been orthogonally by QR decomposition, $U_j^\dagger U_j = I$, we obtain:

$$
\langle \delta X_j | \delta X_k \rangle = \text{Tr}[V_d \cdots V_{j+1} \delta C_j^< U_j^\dagger \cdots U_{k-1} \delta C_k^< V_{k+1}^\dagger \cdots V_d^\dagger] = 0,
$$

(106)

since $\delta C_j^< U_j = 0$. For $j = k$, we obtain:

$$
\langle \delta X_j | \delta X_j \rangle = \text{Tr}[V_d \cdots V_{j+1} \delta C_j^< \delta C_j^< V_{j+1}^\dagger \cdots V_d^\dagger],
$$

$$
= \text{Tr}[\delta C_j^< \delta C_j^<] = \langle \delta C_j^< \delta C_j^< \rangle,
$$

(107)

where, in the second row of Eq. (107), we have used the invariance of the trace with respect to cyclic permutations. Therefore, any $\delta X$ that belongs to the TT tangent space can be written as a direct sum of mutually orthogonal tensors, as follows:

$$
\delta X = \sum_{j=1}^{d} \delta X_j.
$$

(108)

E.2 TT projection operator

Now, we show how the representation of the tangent space $T_Y \mathcal{M}_r$ as the direct sum of mutually orthogonal subsets, introduced by Eqs. (103) and (104), facilitates the derivation of the TT projection operator, introduced by Eq. (14).

Given an arbitrary tensor $Z \in \mathbb{C}^{n_1 \times \cdots \times n_d}$ in TT format (e.g., $Z = M(Y)$), the projection operator $P_Y$ onto the tangent space at $Y$ ensures that $\langle P_Y (Z) - Z | \delta X \rangle = 0$, where $\delta X \in \mathbb{C}^{n_1 \times \cdots \times n_d}$. 


\[ T_Y \mathcal{M}_r, \text{ so} \]
\[ \langle P_Y(Z) | \delta X \rangle = \langle Z | \delta X \rangle. \]  \hfill (109)

Considering that both \( P_Y(Z) \) and \( \delta X \) are in the tangent space, they can both be written in terms of orthogonal decompositions (Eq. (108)), as follows:

\[ P_Y(Z) = \sum_{j=1}^{d} \delta U_j, \]  \hfill (110)

where \( \delta U_j \in \mathcal{V}_j \) and \( \delta X = \sum_{j=1}^{d} \delta X_j \). So, for each mutually orthogonal subspace \( \mathcal{V}_j \), we obtain:

\[ \langle \delta U_j | \delta X_j \rangle = \langle Z | \delta X_j \rangle, \]  \hfill (111)

where \( \delta X_j \in \mathcal{V}_j \) with \( j = 1, \ldots, d \).

Considering that \( X \) is left-orthogonalized, according to Eqs. (90) and (93), \( \delta X_{(j)} = (X \leq j-1 \otimes I_{n_j}) \delta C_{(j)} X_{(j)}^\dagger X_{(j)} \) and \( \delta U_{(j)} = (X \leq j-1 \otimes I_{n_j}) \delta B_{(j)} X_{(j)}^\dagger \). Therefore,

\[ \langle \delta U_j | \delta X_j \rangle = \text{Tr}[(X \leq j-1 \otimes I_{n_j}) \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger (X \leq j-1 \otimes I_{n_j}) \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger], \]
\[ = \text{Tr}[X_{(j)}^\dagger \delta B_{(j)} X_{(j)}^\dagger (X \leq j-1 \otimes I_{n_j}) \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger], \]
\[ = \text{Tr}[X_{(j)}^\dagger X_{(j)}^\dagger \delta B_{(j)} X_{(j)}^\dagger \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger], \]
\[ = \langle \delta B_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger | \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger \rangle. \]  \hfill (112)

Furthermore,

\[ \langle Z | \delta X_j \rangle = \text{Tr}[Z_{(j)}^\dagger (X \leq j-1 \otimes I_{n_j}) \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger], \]
\[ = \text{Tr}[X_{(j)}^\dagger Z_{(j)}^\dagger (X \leq j-1 \otimes I_{n_j}) \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger], \]  \hfill (113)

\[ = \langle (X \leq j-1 \otimes I_{n_j})^\dagger Z_{(j)}^\dagger X_{(j)}^\dagger X_{(j)}^\dagger | \delta C_{(j)} X_{(j)}^\dagger X_{(j)}^\dagger \rangle. \]
Substituting Eqs. (112) and (113) into Eq. (111), we obtain:

$$\langle \delta B_{\leq j+1} X_{\leq j+1} | \delta C_{\leq j+1} \rangle = \langle (X_{\leq j-1} \otimes I_n) | Z^{(j)} X_{\leq j+1} | \delta C_{\leq j} \rangle,$$

which implies that \( \delta B_{\leq j+1} X_{\leq j+1} - (X_{\leq j-1} \otimes I_n) Z^{(j)} X_{\leq j+1} \) is orthogonal to \( \delta C_{\leq j} \). Considering the gauge condition \( U_{\leq j}^\dagger \delta C_{\leq j} = 0 \), the projection operator \( P_{\leq j} = U_{\leq j} U_{\leq j}^\dagger \) onto the range of \( U_{\leq j} \) for \( j = 1, \ldots, d-1 \), and \( P_{\leq d} = 0 \), we obtain:

$$\langle I_j - P_{\leq j} \rangle (\delta B_{\leq j+1} X_{\leq j+1} - (X_{\leq j-1} \otimes I_n) Z^{(j)} X_{\leq j+1}) = 0.$$

Note that Eq. (115) is the tensor-train generalization of Eq. (48).

Rearranging Eq. (115), we obtain:

$$\langle I_j - P_{\leq j} \rangle \delta B_{\leq j} = (I_j - P_{\leq j}) (X_{\leq j-1} \otimes I_n) Z^{(j)} X_{\leq j+1} (X_{\leq j+1}^\dagger X_{\geq j+1})^{-1}.$$

Using the gauge condition \( U_{\leq j}^\dagger \delta B_{\leq j} = 0 \) and the projector \( P_{\leq j} = U_{\leq j} U_{\leq j}^\dagger \), we can show that \( \langle I_j - P_{\leq j} \rangle \delta B_{\leq j} = \delta B_{\leq j} \), since

$$\langle I_j - P_{\leq j} \rangle \delta B_{\leq j} = I_j \delta B_{\leq j} - P_{\leq j} \delta B_{\leq j} = \delta B_{\leq j} - U_{\leq j} U_{\leq j}^\dagger \delta B_{\leq j} = \delta B_{\leq j} - \delta B_{\leq j} = 0,$$

Substituting Eq. (117) into the left hand side of Eq. (116), we obtain:

$$\delta B_{\leq j} = (I_j - P_{\leq j}) (X_{\leq j-1} \otimes I_n) Z^{(j)} X_{\geq j+1} (X_{\geq j+1}^\dagger X_{\geq j+1})^{-1}.$$
Inserting Eq. (118) into $\delta U^{(j)} = (X_{\leq j-1} \otimes I_{n_j}) \delta B^{\leq} X_{\geq j+1}^\dagger$, we obtain:

$$
\delta U^{(j)} = (X_{\leq j-1} \otimes I_{n_j})(I_j - P_j^<)(X_{\leq j-1} \otimes I_{n_j})^\dagger Z^{(j)} X_{\geq j+1}(X_{\geq j+1}^\dagger X_{\geq j+1})^{-1} X_{\geq j+1}^\dagger. \tag{119}
$$

Introducing the following projectors,

$$
P_{\leq j-1} = X_{\leq j-1} X_{\leq j-1}^\dagger,
$$

$$
P_{\leq i} = (X_{\leq j-1} \otimes I_{n_j}) P_j^< (X_{\leq j-1} \otimes I_{n_j})^\dagger, \tag{120}
$$

$$
P_{\geq j+1} = X_{\geq j+1}(X_{\geq j+1}^\dagger X_{\geq j+1})^{-1} X_{\geq j+1}^\dagger,
$$

we simplify Eq. (119), as follows:

$$
\delta U^{(j)} = (P_{\leq j-1} \otimes I_{n_j}) Z^{(j)} P_{\geq j+1}, \quad j = 1, \ldots, d - 1
$$

$$
\delta U^{(d)} = (P_{\leq d-1} \otimes I_{n_d}) Z^{(d)}. \tag{121}
$$

Note that $\delta U^{(d)}$ as defined in Eq. (121) has only one term since $P_d^< = 0$, so $P_{\leq d} = 0$.

Considering that according to Eq. (110), $P_Y(Z) = \sum_{j=1}^d \delta U_j$, we obtain:

$$
P_Y(Z) = \sum_{j=1}^{d-1} \text{Ten}_j[(P_{\leq j-1} \otimes I_{n_j}) Z^{(j)} P_{\geq j+1} - P_{\leq j} Z^{(j)} P_{\geq j+1}] + \text{Ten}_d[(P_{\leq d-1} \otimes I_{n_d}) Z^{(d)}]. \tag{122}
$$

To simplify the notation, we define:

$$
P^+_j(Z) = \text{Ten}_j[(P_{\leq j-1} \otimes I_{n_j}) Z^{(j)} P_{\geq j+1}],
$$

$$
P^-_j(Z) = \text{Ten}_j[P_{\leq j} Z^{(j)} P_{\geq j+1}], \tag{123}
$$

we obtain

$$
P_Y(Z) = P^+_1(Z) - P^-_1(Z) + P^+_2(Z) - P^-_2(Z) + \cdots - P^-_{d-1}(Z) + P^+_d(Z). \tag{124}
$$
F TT-KSL Integration

F.1 TT-KSL equations of motion

The TT-KSL method integrates the equations of motion for $Y^+_i(t)$ and $Y^-_i(t)$, introduced by Eq. (16), sweeping from left to right to update the cores of the tensor $Y(t_0)$.

The initial state $Y_0$ is right orthogonalized at core 1, as follows (Appendix D.5):

$$\begin{align*}
[Y_1^+(t_0)]^{(1)} &= Y_{\leq 1}(t_0)R_2^\dagger(t_0)V_{\geq 2}(t_0), \\
&= K_1^<(t_0)V_{\geq 2}(t_0),
\end{align*}$$

where $Y_{\leq 1}(t_0)$ is core 1 of $Y_0$, and $K_1^<(t) = Y_{\leq 1}(t)R_2^\dagger(t_0)$. With the right-orthogonalized $Y_1^+(t_0)$, we solve the first equation in Eq. (16), written as the unfolding matrix equation:

$$\begin{align*}
[\dot{Y}_1^+(t)]^{(1)} &= [P_1^+(MY_1^+(t))]^{(1)}.
\end{align*}$$

We integrate Eq. (126) with $V_{\geq 2}$ fixed in time, analogous to the first step of the KSL integration for matrices (Appendix C.1), as follows:

$$\begin{align*}
[\dot{Y}_1^+(t)]^{(1)} &= \dot{K}_1^<(t)V_{\geq 2}(t_0),
\end{align*}$$

which gives

$$\begin{align*}
[Y_1^+(t_1)]^{(1)} &= K_1^<(t_1)V_{\geq 2}(t_0).
\end{align*}$$

We obtain $K_1^<(t_1)$ by integrating the equation of motion for $K_1^<(t)$ (i.e., Eq. (131)), which can be obtained, as follows. We substitute Eq. (123) into Eq. (126), as follows:

$$\begin{align*}
[P_1^+(MY_1^+(t))]^{(1)} &= P_{\leq 0}[MY_1^+(t)]^{(1)}P_{\geq 2},
\end{align*}$$

where $P_{\leq 0} = 1$. Next, we substitute the projection operators in Eq. (129), according to
Eq. (120), and we obtain:

\[
P^+_1(MY^+_1(t))^{(1)} = [MY^+_1(t)]^{(1)}V_{\geq 2}(V^\dagger_{\geq 2}V_{\geq 2})^{-1}V^\dagger_{\geq 2},
\]

\[
= [MY^+_1(t)]^{(1)}V_{\geq 2},
\]  

(130)

where the second row is obtained with \(V^\dagger_{\geq 2}V_{\geq 2} = I\). Finally, we equate the right-hand-sides of Eqs. (126) and (127) and substitute \([P^+_1(MY^+_1(t))]^{(1)}\) according to Eq. (130), to obtain:

\[
\dot{K}^<_1(t) = [MY^+_1(t)]^{(1)}V_{\geq 2}(t_0).
\]  

(131)

\(K^<_1(t_1)\) is obtained by integration of Eq. (131) from \(t_0\) to \(t_1\) and substituted into Eq. (128) to obtain \(Y^+_1(t_1)\). In general, Eq. (131) can be integrated by the Runge-Kutta method. In applications to model systems where \(M\) is time-independent, such as in the integration of Eq. (12) for the time-independent Hamiltonian of rhodopsin introduced by Eq. (1), Eq. (131) corresponds to a constant-coefficient ordinary differential equation (ODE) that can be integrated by computing the action of a matrix exponential in the Krylov subspace (i.e., Eq. (159), Sec. [F.3]), as implemented in Expokit.\footnote{79}

Having obtained \(Y^+_1(t_1)\), we complete the propagation of core 1 by integrating \(\dot{Y}^-_1 = P^-_1(MY^-_1)\) with initial condition \(Y^-_1(t_0) = Y^+_1(t_1)\). We start by orthogonalizing the first core of \(Y^-_1(t_0)\), according to the QR decomposition \(K^<_1(t_1) = U^<_1(t_1)R^<_1(t_1)\), as follows:

\[
[Y^-_1(t_0)]^{(1)} = [Y^+_1(t_1)]^{(1)},
\]

\[
= K^<_1(t_1)V^\dagger_{\geq 2}(t_0),
\]

\[
= U^<_1(t_1)R^<_1(t_1)V^\dagger_{\geq 2}(t_0),
\]

\[
= U^<_1(t_1)S_1(t_0)V^\dagger_{\geq 2}(t_0),
\]

(132)

where we have introduced the substitution \(S_1(t_0) = R^<_1(t_1)\). The fourth equality comes
from the recursive construction relationship (Eq. (93)), which is trivial for $i = 1$, and useful for subsequent substeps. Analogous to the second step of the KSL integration for matrices (Appendix C.2), we evolve

$$[Y_1^{-}(t)]^{(1)} = U_{\leq 1}(t_1)S_1(t)\mathbf{V}_2^\dagger(t_0),$$

(133)

from $t_0$ to $t_1$, by keeping $U_{\leq 1}(t_1)$ fixed and requiring $\mathbf{V}_{\geq 2} = \mathbf{0}$, such that only $S_1$ is allowed to change over time during the propagation. Therefore, $[Y_1^{-}(t)]^{(1)}$ evolves, as follows:

$$[\dot{Y}_1^{-}(t)]^{(1)} = U_{\leq 1}(t_1)\frac{dS_1(t)}{dt}\mathbf{V}_{\geq 2}^\dagger(t_0).$$

(134)

We obtain the equation of motion for $\dot{S}_1(t)$, as follows. We substitute $P_1^{-}$ according to Eq. (123),

$$[\dot{Y}_1^{-}(t)]^{(1)} = [P_1^{-}(MY_1^{-}(t))]^{(1)},$$

$$= P_{\leq 1}[MY_1^{-}(t)]^{(1)}P_{\geq 2},$$

$$= U_{\leq 1}U_{\leq 1}^\dagger[MY_1^{-}(t)]^{(1)}\mathbf{V}_{\geq 2}(\mathbf{V}_{\geq 2}^\dagger\mathbf{V}_{\geq 2})^{-1}\mathbf{V}_{\geq 2}^\dagger,$$

$$= U_{\leq 1}U_{\leq 1}^\dagger[MY_1^{-}(t)]^{(1)}\mathbf{V}_{\geq 2}\mathbf{V}_{\geq 2}^\dagger,$$

(135)

where $U_{\leq 1}$ and $\mathbf{V}_{\geq 2}$ are kept fixed and $\mathbf{V}_{\geq 2}$ is orthogonalized. Comparing Eqs. (135) and (134), we obtain the equation of motion for $S_1(t)$, as follows:

$$\frac{dS_1(t)}{dt} = U_{\leq 1}(t_1)[MY_1^{-}(t)]^{(1)}\mathbf{V}_{\geq 2}(t_0).$$

(136)

$S_1(t)$ is obtained by integrating Eq. (136) with Expokit\textsuperscript{79} and substituted into Eq. (133) to obtain $[Y_1^{-}(t_1)]^{(1)}$, as follows:

$$[Y_1^{-}(t_1)]^{(1)} = U_{\leq 1}(t_1)S_1(t_1)\mathbf{V}_{\geq 2}^\dagger(t_0).$$

(137)
Having updated the first core, we proceed with the sweeping method to update the second core according to the next two equations of motion introduced by Eq. (16), which involve $P_{2}^{+}$ and $P_{2}^{-}$. The initial state $[Y_{2}^{+}(t_{0})]^{(2)} = [Y_{1}^{-}(t_{1})]^{(2)}$ is obtained by refolding $[Y_{1}^{-}(t_{1})]^{(1)}$ into $[Y_{1}^{-}(t_{1})]^{(2)}$, as follows:

$$[Y_{2}^{+}(t_{0})]^{(2)} = [\text{Ten}_1[Y_{1}^{-}(t_{1})]^{(1)}]^{(2)},$$

$$= [\text{Ten}_1[U_{\leq 1}(t_{1})S_{1}(t_{1})V_{\geq 2}^{\dagger}(t_{0})]]^{(2)},$$

$$= (I_{n_{1}} \otimes U_{\leq 1}(t_{1}))(I_{n_{1}} \otimes S_{1}(t_{1}))V_{2}^{\leq}(t_{0})V_{\geq 3}^{\dagger}(t_{0}),$$

where the last equality used Eqs. (101) and (102). Absorbing $S_{1}(t_{1})$ into core 2 and defining $K_{2}^{\leq}(t_{0}) = (I_{n_{1}} \otimes S_{1}(t_{1}))V_{2}^{\leq}(t_{0})$, we obtain:

$$[Y_{2}^{+}(t_{0})]^{(2)} = (I_{n_{1}} \otimes U_{\leq 1}(t_{1}))K_{2}^{\leq}(t_{0})V_{\geq 3}^{\dagger}(t_{0}).$$

(139)

The propagation of Eq. (139) follows the same procedure as described for Eq. (125) to generate $[Y_{2}^{+}(t_{1})]^{(2)}$. $[Y_{2}^{-}(t_{0})]^{(2)}$ is initialized by $[Y_{2}^{+}(t_{1})]^{(2)}$ and propagated, as described for Eq. (132), to generate $[Y_{2}^{-}(t_{1})]^{(2)}$. The same procedure is sequentially applied to update core-by-core all cores of the tensor train – i.e., the so-called ‘sweeping update’ algorithm.

### F.2 Sweeping algorithm

The sweeping algorithm for updating cores $i = 2, \ldots, d$, implements the procedure applied for updating the first core introduced in Sec. F.1, which requires integration of the equation of motion, $\dot{Y}_{i}^{+}(t) = P_{i}^{+}(MY_{i}^{+}(t))$, for the time interval $[t_{0}, t_{1}]$. The equation is written as the unfolding matrix equation,

$$[\dot{Y}_{i}^{+}(t)]^{(i)} = [P_{i}^{+}(MY_{i}^{+}(t))]^{(i)},$$

(140)
with initial conditions $Y_{i}^{+}(t_0) = Y_{i-1}^{-}(t_1)$. These quantities are left- and right- orthogonalized in terms of the previously updated core, as described by Eq. (139) for $Y_{2}^{+}(t_0)$:

$$[Y_{i}^{+}(t_0)]^{(i)} = (U_{\leq i-1}(t_1) \otimes I_{n_i})K_{i}^{<}(t_0)V_{\geq i+1}^{\dagger}(t_0), \tag{141}$$

with $V_{\geq d+1} = 1$. Eq. (141) is true for $i = 2$, as shown by Eq. (139), and can be shown to be valid for any $i$ by induction.

We update $[Y_{i}^{+}(t)]^{(i)}$ with constant $U_{\leq i-1}$ and $V_{\geq i+1}^{\dagger}$, by propagating $K_{i}^{<}(t)$ and substituting into Eq. (141), as follows:

$$[Y_{i}^{+}(t)]^{(i)} = (U_{\leq i-1}(t_1) \otimes I_{n_i})K_{i}^{<}(t)V_{\geq i+1}^{\dagger}(t_0). \tag{142}$$

The equation of motion for $K_{i}^{<}(t)$ is obtained from the explicit time derivative of $[Y_{i}^{+}(t)]^{(i)}$, as defined by Eq. (142), as follows we obtain:

$$[\dot{Y}_{i}^{+}(t)]^{(i)} = [P_{i}^{+}(MY_{i}^{+})(t)]^{(i)},$$

$$= \frac{d}{dt}[(U_{\leq i-1}(t_1) \otimes I)K_{i}^{<}(t)V_{\geq i+1}^{\dagger}(t_0)], \tag{143}$$

$$= (U_{\leq i-1}(t_1) \otimes I_{n_i})\frac{d}{dt}[K_{i}^{<}(t)]V_{\geq i+1}^{\dagger}(t_0).$$

where, according to Eq. (123),

$$[P_{i}^{+}(MY_{i}^{+})(t)]^{(i)} = (P_{\leq i-1} \otimes I_{n_i})[MY_{i}^{+}(t)]^{(i)}P_{\geq i+1}. \tag{144}$$

Substituting the projection operators in Eq. (144), according to Eq. (120), we obtain:

$$[P_{i}^{+}(MY_{i}^{+})(t)]^{(i)} = ((U_{\leq i-1}(t_1)U_{\leq i-1}(t_1)) \otimes I_{n_i})[MY_{i}^{+}(t)]^{(i)}V_{\geq i+1}(t_0)V_{\geq j+1}^{\dagger}(t_0),$$

$$= (U_{\leq i-1}(t_1) \otimes I_{n_i})(U_{\leq i-1}(t_1) \otimes I_{n_i})[MY_{i}^{+}(t)]^{(i)}V_{\geq i+1}(t_0)V_{\geq j+1}^{\dagger}(t_0). \tag{145}$$
and substituting Eq. (145) into Eq. (143), we obtain:

\[
(U \leq i-1(t_1) \otimes I_{m_i}) \frac{d}{dt} [K_i^<(t)] V_{\geq j+1}(t_0) = (U \leq i-1(t_1) \otimes I_{m_i})(U \leq i-1(t_1) \otimes I_{m_i})^\dagger [MY_i^+(t)]^{(i)} V_{\geq j+1}(t_0) V_{\geq j+1}(t_0).
\]

(146)

Simplifying Eq. (146), we obtain:

\[
\dot{K}_i^< = (U \leq i-1(t_1) \otimes I_{m_i}) [MY_i^+(t)]^{(i)} V_{\geq i+1}(t_0).
\]

(147)

Considering that \(U \leq i-1(t)\) and \(V_{\geq i+1}(t)\) are held fixed in time, we can update \(Y_i^+\), as defined by Eq. (142), by updating only core \(i\) while keeping all other cores unchanged, as follows:

\[
[Y_i^+(t_1)]^{(i)} = (U \leq i-1(t_1) \otimes I_{m_i}) K_i^<(t_1) V_{\geq i+1}(t_0),
\]

(148)

or in matrix product notation,

\[
Y_i^+(j_1, \ldots, j_d, t_1) = U_1(j_1, t_1) \ldots U_{i-1}(j_{i-1}, t_1) K_i(j_i, t_1) V_{i+1}(j_{i+1}, t_0) \ldots V_d(j_d, t_0).
\]

(149)

Having obtained \(Y_i^+(t_1)\), we complete the propagation of core \(i\) by integrating \(\dot{Y}_i^- = P_i^- (MY_i^-)\) with initial condition \(Y_i^-(t_0) = Y_i^+(t_1)\). Similar to the process in Eq. (132), we orthogonalize the \(i^{th}\) core by substituting \(K_i^<(t_1)\) in Eq. (148) according to the QR
decomposition $K^<_i(t_1) = U^<_i(t_1)R_i(t_1)$, as follows:

$$[Y^+_i(t_0)]^{(i)} = [Y^+_i(t_1)]^{(i)};$$

$$= (U_{\leq i-1}(t_0) \otimes I_{n_i})K^<_i(t_1)V^\dagger_{\geq i+1}(t_0);$$

$$= (U_{\leq i-1}(t_1) \otimes I_{n_i})U^<_i(t_1)R_i(t_1)V^\dagger_{\geq i+1}(t_0);$$

$$= (U_{\leq i-1}(t_1) \otimes I_{n_i})U^<_i(t_1)R_i(t_1)V^\dagger_{\geq i+1}(t_0);$$

$$Y^-_i(t_0) = \langle i \rangle;$$

$$= (U_{\leq i}(t_1) \otimes I_{n_i})S_i(t_0)S^\dagger_{i+1}(t_0).$$

(150)

where we have introduced the substitution $S_i(t_0) = R_i(t_1)$. Since Eq. (150) is analogous to Eq. (137) (where $i = 1$), updating $Y^-_i(t)$ requires the same process as applied to $Y^-_1(t)$ (Eq. (134) to (139)). Analogous to Eq. (138), we obtain:

$$[Y^-_i(t_1)]^{(i+1)} = (I_{n_i} \otimes U_{\leq i}(t_1))(I_{n_i} \otimes S_i(t_1))V^\dagger_{i+1}(t_0)V^\dagger_{\geq i+2}(t_0);$$

(151)

where $S_i(t_1)$ is obtained by integrating the following equation:

$$\dot{S}_i(t) = U^\dagger_{\leq i}[MY^-_i(t)]^{(i)}V^\dagger_{\geq i+1}. $$

(152)

Having obtained $K^<_i(t_0) = S_i(t_1))V^\dagger_{i+1}(t_0)$, we initialize $Y^+_i$ as in Eq. (142), as follows:

$$[Y^+_i(t)]^{(i+1)} = (U_{\leq i}(t) \otimes I_{n_i+1})K^<_i(t)V^\dagger_{\geq i+2}(t).$$

(153)

Updating all cores, according to Eqs. (142)–(153), yields $Y^+_d(t_1)$, which that approximates the propagated tensor, as follows $Y(t_1) = Y^+_d(t_1)$, since according to Eq. (16), the $d^{th}$ core involves only $Y^+_d$.

As mentioned in Sec. F.1 for the propagation of the first core, the equations of motion
introduced by Eqs. (147) and (152) can be integrated by the Runge-Kutta method. Furthermore, in applications to model systems where $M$ is time-independent as in the our application to rhodopsin, those equations can be integrated by computing the action of a matrix exponential in the Krylov subspace with Expokit, as shown in the Sec. F.3 (Eq. (159)).

F.3 Integration with matrix exponential

This section shows that the right-hand side of Eq. (147), with a time-independent $M$, defines a constant-coefficient ODE with respect to $K^<_{i}(t)$, an equation that can be integrated by computing the action of a matrix exponential. Analogously, we could show that Eq. (152) defines a constant-coefficient ODE with respect to $S_i(t)$, such that both equations of motion can be integrated analogously.

$MY^+_{i}(t)$ can be written by using Eqs. (149) and (6), as follows:

$$MY^+_{i}(l_1,...,l_d;t) = \sum_{j_i} M_1(l_1,j_1) \otimes U_1(j_1;t_0) \cdots \sum_{j_i-1} M_{i-1}(l_i-1,j_i-1) \otimes U_{i-1}(j_{i-1};t_0)$$

$$\left( \sum_{j_i} M_i(l_i,j_i) \otimes K_i(j_i;t) \right) \left( \sum_{j_{i+1}} M_{i+1}(l_{i+1},j_{i+1}) \otimes V_{i+1}(j_{i+1};t_0) \right) \cdots$$

$$\left( \sum_{j_d} M_d(l_d,j_d) \otimes U_d(j_d;t_0) \right),$$

which can be written in the unfolding matrix format, as follows:

$$[MY^+_{i}(t)](i) = (I_n \otimes \bar{U}_{\leq i-1}(t_0)) \bar{K}_{i}^<(t) \bar{V}_{\geq i+1}^t(t_0),$$

where $\bar{U}_{\leq i-1}$ represents the left-unfolding matrices for the first $i-1$ cores and $\bar{V}_{\geq i+1}$ represents the right-unfolding matrices for the last $d-i$ cores of Eq. (154). $\bar{K}_{i}^<(t)$ corresponds to the $i^{th}$ core, and is obtained by operating $M_i$ on $K_{i}^<(t)$ with proper reshaping.
Inserting Eq. (155) into Eq. (147) yields:

\[
\frac{d}{dt}[K_i^<(t)] = (I_{n_i} \otimes U_{\leq i-1}^\dagger(t_0))(I_{n_i} \otimes \bar{U}_{\leq i-1}(t_0))K_i^<(t)\bar{V}_{\geq i+1}(t_0)V_{\geq i+1}(t_0),
\]  

which has time dependency only for core \( i \).

Defining \( W_iK_i^<(t) \), as follows:

\[
W_iK_i^<(t) = (I_{n_i} \otimes U_{\leq i-1}^\dagger(t_0))(I_{n_i} \otimes \bar{U}_{\leq i-1}(t_0))K_i^<(t)\bar{V}_{\geq i+1}(t_0)V_{\geq i+1}(t_0),
\]  

we rewrite Eq. (147), as follows:

\[
\frac{d}{dt}[K_i^<(t)] = W_i[K_i^<(t)].
\]  

Considering that \( W_i \) is time-independent, Eq. (158) can be formally integrated, as follows:

\[
K_i^<(t_1) = e^{(t_1-t_0)W_i}K_i^<(t_0),
\]  

and the matrix exponential can be numerically computed core-by-core by using the Krylov space method, as implemented in the EXPOKIT package.\textsuperscript{23}

\section*{G Codes for quantum dynamics simulations}

The Python codes for TT-SOKSL, TT-KSL, TT-SOFT, and full-grid SOFT simulations of retinal model are available at: https://github.com/NingyiLyu/TTSOKSL.

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Figure 10: Table of contents (TOC).